

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property
Organization
International Bureau



(43) International Publication Date
8 April 2004 (08.04.2004)

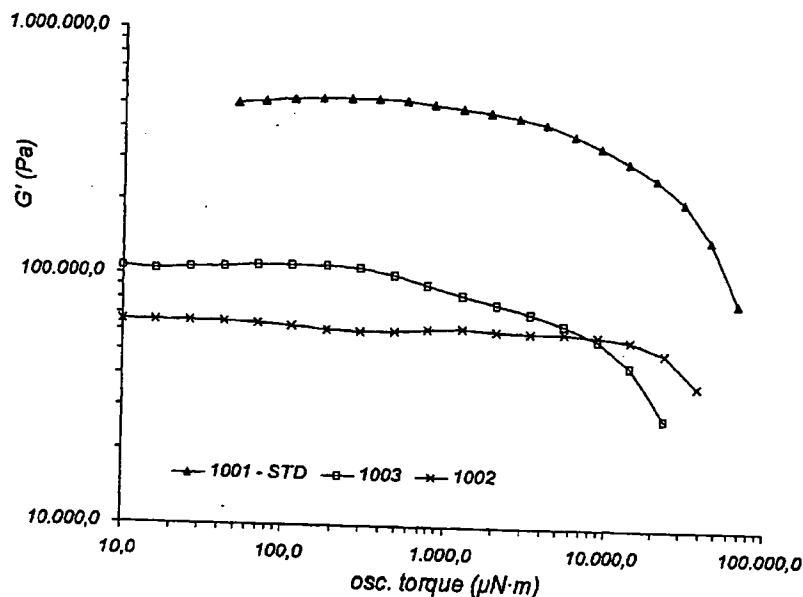
PCT

(10) International Publication Number
WO 2004/028269 A1

- (51) International Patent Classification⁷: A23G 3/30 // C08G 63/08, 63/64, C08L 67/04 (74) Agent: PATENTGRUPPEN APS; Arosgården, Åboulevarden 31, DK-8000 Århus C (DK).
- (21) International Application Number: PCT/DK2002/000628 (81) Designated States (*national*): AE, AG, AL, AM, AT (utility model), AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ (utility model), CZ, DE (utility model), DE, DK (utility model), DK, DM, DZ, EC, EE (utility model), EE, ES, FI (utility model), FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, OM, PH, PL, PT, RO, RU, SD, SE, SG, SI, SK (utility model), SK, SL, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZM, ZW.
- (22) International Filing Date: 24 September 2002 (24.09.2002) (84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, SK, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).
- (25) Filing Language: English
- (26) Publication Language: English
- (71) Applicant (*for all designated States except US*): GUM-LINK A/S [DK/DK]; Dandyvej 19, DK-7100 Vejle (DK).
- (72) Inventors; and
- (75) Inventors/Applicants (*for US only*): ANDERSEN, Lone [DK/DK]; Gl. Strandvej 1, DK-5500 Middelfart (DK). WITTORE, Helle [DK/DK]; Hegnsgårdsvej 63, DK-7120 Vejle Ø (DK). STOREY, Robson [US/US]; 111 Holly Dr., Hattiesburg, MS 39402 (US). DESAI, Ganesh, S. [IN/US]; The University of Southern Mississippi, Department of Polymer Science, USM Box - 10076, Hattiesburg, MS 39406 (US).
- Published:
— with international search report

[Continued on next page]

(54) Title: DEGRADABLE CHEWING GUM POLYMER



(57) Abstract: The invention relates to degradable chewing gum polymer, said degradable polymer is a polymer polymerized from at least one trifunctional or higher functional initiator, at least two different monomers forming the backbone of the polymer and at least one monomer selected from the group of carbonate monomers. According to the invention it has been realized that a certain degree of branching of the backbone is needed to obtain a final improved performance, when the polymer, preferably the elastomer, is incorporated in a chewing gum. It has moreover been realized that the obtained degree of branching needs and may actually be carefully controlled in order to avoid too much branching-induced crosslinking.

BEST AVAILABLE COPY

WO 2004/028269 A1



For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

DEGRADABLE CHEWING GUM POLYMER

Field of the invention

- 5 The present invention relates to a degradable chewing gum polymer according to claim 1.

Background of the invention

- 10 US patent 5,672,367 discloses a biodegradable elastomer for chewing gum. The elastomers are generally defined as biodegradable polyester polymers obtained by the polymerization of one or more cyclic esters. Two specific examples are described.
- 15 Example 1 describes an amorphous, non-crystallizable copolymer of a polymer of 80 mol % L-lactide and 20 mol % D-lactide that was prepared by ring-opening polymerization in the melt, in the presence of 0,1% by weight tin octoate as a catalyst. To this polymer was added an amount of 20% by weight of epsilon-caprolactone, and subsequently the mixture was heated to 150°C. To the
- 20 homogeneous mixture, again 0,1% by weight tin octoate as catalyst was added and then the polymerization was completed. The obtained polymer had a glass transition temperature (DSC, heating rate 10°C/min) of 15°C.

- Example 3 describes an amorphous, non-crystallizable copolymer of 25 mol % L-lactide, 25 mol % D-lactide and 50 mol % epsilon-caprolactone that was prepared by
- 25 ring-opening polymerization in the melt, in the presence of 0,1% by weight tin octoate as catalyst. The obtained polymer has a glass transition temperature (DSC, heating rate 10°C/min) of -10°C

- 30 Both exemplified polymers is stated to feature a chew feel strongly resembling that of conventional chewing gum.

However, a disadvantage of the above mentioned polymers is that the properties of the provided polymers differ from conventional chewing gum elastomers for example with respect to the texture of the polymers itself and especially when incorporated in conventional chewing gum formulations.

5

WO 01/47368 discloses a chewing gum comprising a degradable copolymer obtained by polymerization of two different monomers, one first monomer which is polymerizable by condensation polymerization and one monomer functional to suppress the crystallinity of the copolymer. A problem of the disclosed copolymer is however for example that the elastomeric properties of the resulting copolymer differ when compared to properties of conventional chewing gum. Consequently, it appears very difficult to obtain a completely biodegradable chewing gum based on the disclosed copolymer illustrated by the fact that the examples only disclose partly biodegradable chewing gum.

15

It is an object of the invention to provide a chewing gum polymer having properties comparable to those of conventional chewing gum elastomers both with respect to the polymer itself and with respect to the interaction with the chewing gum ingredients when incorporated in a chewing gum formulation.

20

Summary of the invention

The invention relates to a degradable chewing gum polymer,
25 said degradable polymer being a polymer polymerized from

at least one trifunctional or higher functional initiator

at least two different monomers forming the backbone of the polymer and

30

at least one monomer selected from the group of carbonate monomers.

According to the invention, the obtained polymer has elastomeric properties suitable for chewing gum.

According to the invention, a polymer structure being very suitable as chewing
5 polymer/elastomer has been obtained.

According to the invention it has been realized that a certain degree of branching of the backbone is needed to obtain a final improved performance, when the polymer, preferably the elastomer, is incorporated in a chewing gum. It has moreover been
10 realized that the obtained branching needs to be carefully controlled in order to avoid too much branching-induced crosslinking.

According to the invention, it has surprisingly been realized that this balance between branching/cross-linking may be controlled by a suitable pairing of initiator
15 and carbonate monomer. Such pairing includes among the most significant "control knobs" the mutual concentration of the initiator versus the carbonate monomer.

Moreover, the mutual concentration may be modified under consideration of the structure of the initiator. The higher functional initiator, the lower concentration of
20 the carbonate monomer.

According to the invention, the term hyperbranched preferably indicates that the branching structure is dendritic rather than comb-like. That is, branches extend from other branches, like a tree, rather than many simple branches extending from a well-
25 defined backbone segment (comb-like branching). Hence, hyperbranching may be understood as "branching of a dendritic nature." Branching in this system is an intermediate stage leading to crosslinking. The molecules first become branched, and then when a branch from one molecule reacts with a branch of another molecule, a crosslink is formed. At intermediate stages within this process, branched and
30 crosslinked molecules coexist. The man of ordinary skill in the art will understand branching and crosslinking and the difference between dendritic and comb-like branching. A good description of dendritic branching compared to other types of

branching can be found in the following textbook:

Odian, G. "Principles of Polymerization," 3rd Ed., Wiley-Interscience, New York, NY (1991); p. 17.

5

Preferably said at least two different monomers are cyclic.

In an embodiment of the invention the at least two different monomers forming the backbone of the polymer comprise at least one backbone monomer and a at least one backbone comonomer.

10

In an embodiment of the invention the at least one backbone comonomer imparts disorder in the backbone monomer chain.

According to the invention, it has been realized that the backbone chain comprises at least two different monomers.

15

In an embodiment of the invention the at least one backbone comonomer is effective to introduce amorphous regions in the backbone monomer chain.

20

In an embodiment of the invention the at least two different monomers forming the backbone of the polymer are selected from the group of lactone monomers.

25

In an embodiment of the invention the lactone monomers are chosen from the group of ϵ -caprolactone, δ -valerolactone, γ -butyrolactone, and β -propiolactone. It also includes ϵ -caprolactones, δ -valerolactones, γ -butyrolactones, or β -propiolactones that have been substituted with one or more alkyl or aryl substituents at any non-carbonyl carbon atoms along the ring, including compounds in which two substituents are contained on the same carbon atom.

30

Examples of the lactones described above are, but not limited to, -caprolactone, t-butyl caprolactone, zeta-enantholactone, deltavalerolactones, the monoalkyl-delta-

valerolactones, e. g. the monomethyl-, monoethyl-, monohexyl-deltavalerolactones, and the like; the nonalkyl, dialkyl, and trialkyl-epsilon-caprolactones, e. g. the monomethyl-, monoethyl-, monohexyl-, dimethyl-, di-n-propyl-, di-n-hexyl-, trimethyl-, triethyl-, tri-n-epsilon-caprolactones, 5-nonyloxepan-2-one, 4, 4, 6- or 4, 5, 6, 6-trimethyl-oxepan-2-one, 5-hydroxymethyloxepan-2-one, and the like; beta-lactones, e. g., beta-propiolactone, beta-butyrolactone gamma-lactones, e. g., gammabutyrolactone or pivalolactone, dilactones, e. g. lactide, dilactides, glycolides, e. g., tetramethyl glycolides, and the like, ketodioxanones, e. g. 1, 4-dioxan-2-one, 1, 5-dioxepan-2-one, and the like. The lactones can consist of the optically pure isomers or two or more optically different isomers or can consist of mixtures of isomers.

In an embodiment of the invention the at least one backbone monomer comprises ϵ -caprolactone

According to a preferred embodiment of the invention ϵ -caprolactone is chosen as the main monomer of the backbone, thereby ensuring that the main component of the backbone features a sufficiently low Tg.

In an embodiment of the invention the at least one backbone monomer has a Tg below -40°C , preferably less than -50°C .

In an embodiment of the invention the at least one backbone comonomer comprises δ -valerolactone.

According to a preferred embodiment of the invention δ -valerolactone forms a suitable backbone comonomer. Moreover, it has been realized that the requirements with respect to a low Tg may be somewhat relaxed, when compared to the constraints on the main backbone monomer.

Evidently, it should be noted that the Tg of the comonomer or comonomers becomes more significant with increasing concentration.

In an embodiment of the invention said degradable polymer is polymerized by metal catalyzed ring-opening.

Preferably the carbonate monomer is selected from the group of trimethylene carbonate, 5-alkyl-1,3-dioxan-2-one, 5,5-dialkyl-1,3-dioxan-2-one, or 5-alkyl-5-alkyloxycarbonyl-1,3-dioxan-2-one.

Examples of suitable cyclic carbonates are ethylene carbonate, 3-ethyl-3-hydroxymethyl trimethylene carbonate, propylene carbonate, trimethylene carbonate, trimethylolpropane monocarbonate, 4,6-dimethyl-1,3-propylene carbonate, 2,2-dimethyl trimethylene carbonate, and 1,3-dioxepan-2-one and mixtures thereof.

According to the invention several different carboner monomers may be applied. The preferred carbonate monomer is trimethylene carbonate (TMC).

15

In an embodiment of the invention the at least one monomer selected from the group of carbonate monomers provides a means for introducing additional branching and/or crosslinking to the elastomeric polymer during ring-opening polymerization.

According to the invention cyclic carbonate in the monomer mixture yields precise control over the degree of branching and crosslinking in the final polymer. The mechanism by which the cyclic carbonate monomer imparts crosslinking is based upon the known tendency for metal catalysts, of which stannous octoate is a non-limiting example, to promote transesterification and transcarbonation reactions (intermolecular chain transfer to polymer) during polymerization.

25

In an embodiment of the invention said at least one polyol comprises a trifunctional or higher functional initiator.

According to the invention, the interaction between the polyol initiator and the carbonate monomer provides the desired branching of the resulting biodegradable polymer.

30

Another aspect of the present invention is directed to the production of star polymers.

5 Examples of advantageous multifunctional initiators are, but not limited to glycerol, trimethylolpropane, pentaerythritol, dipentaerythritol, ethoxylated or propoxylated polyamines and other molecules with multiple hydroxyl or other reactive groups and other molecules with multiple hydroxyl or other reactive groups and mixtures thereof.

10 According to a preferred embodiment of the invention, the preferred initiators are trimethylolpropane and pentaerythritol.

In an embodiment of the invention the degradable chewing gum polymer is polymerized from:

15

about 20 to 80 wt % of the at least one backbone monomer,
about 19.5 to 79.5 wt % of the at least one backbone comonomer,
about 0.5 to 25 wt % of the at least one monomer selected from the group of carbonate monomers.

20

In an embodiment of the invention the degradable chewing gum polymer is moreover polymerized from:

About 0.01 to 1.0 wt % of the at least one initiator

25

In an embodiment of the invention the chewing gum properties of the polymer are adjusted by selection of a suitable order of the multifunctional initiator.

30 The more functional initiator, the less carbonate for the purpose of generating the desired amount of hyperbranching and crosslinking.

In an embodiment of the invention the rheological properties of the degradable polymer are controlled by adjusting the functional number of initiators.

5 Moreover, it has been realized that an increase in the functionality of the initiator results in an improved texture and/or improved release of chewing gum ingredients when the polymer is incorporated in a chewing gum.

The molecular weight of lactone monomer must be within the range of 50-16000 g/mol preferably within the range of 100-3000 g/mol

10

The molecular weight of carbonate monomer must be within the range 50-15000 g/mol preferably within the range of 100-2300 g/mol.

15 In an embodiment of the invention said chewing gum ingredients comprise flavoring agents.

In an embodiment of the invention said flavoring agents comprise natural and synthetic flavourings in the form of natural vegetable components, essential oils, essences, extracts, powders, including acids and other substances capable of affecting

20

the taste profile

25

In an embodiment of the invention said chewing gum comprises flavor in an amount of 0.01 to about 30 wt %, said percentage being based on the total weight of the chewing gum

25

In an embodiment of the invention said chewing gum comprises flavor in an amount of 0.2 to about 4 wt %, said percentage being based on the total weight of the chewing gum

30 In an embodiment of the invention said flavor comprises water soluble ingredients.

In an embodiment of the invention said water soluble flavor comprises acids.

According to the invention, a surprising initial release of acids has been obtained.

In an embodiment of the invention said flavor comprising water insoluble
5 ingredients.

In an embodiment of the invention, said chewing gum ingredients comprising
sweeteners.

10 In an embodiment of the invention said sweetener comprises bulk sweeteners

In an embodiment of the invention the chewing gum comprises bulk sweeteners in an
amount of about 5 to about 95% by weight of the chewing gum, more typically about
20 to about 80% by weight of the chewing gum.

15 In an embodiment of the invention the sweetener comprises high intensity sweeteners

In an embodiment of the invention the high intensity sweeteners comprises sucralose,
aspartame, salts of acesulfame, alitame, saccharin and its salts, cyclamic acid and its
20 salts, glycyrrhizin, dihydrochalcones, thaumatin, monellin, sterioside, alone or in
combination

In an embodiment of the invention wherein the chewing gum comprises high
intensity sweeteners in an amount of about 0 to about 1% by weight of the chewing
25 gum, more typically about 0.05 to about 0.5 % by weight of the chewing gum.

In an embodiment of the invention, the chewing gum comprises at least one softener.

In an embodiment of the invention, the at least one softener comprises tallow,
30 hydrogenated tallow, hydrogenated and partially hydrogenated vegetable oils, cocoa
butter, glycerol monostearate, glycerol triacetate, lecithin, different waxes, mono-,

di- and triglycerides, acetylated monoglycerides, fatty acids - such as stearic, palmitic, oleic and linoleic acids mixtures thereof.

In an embodiment of the invention the chewing gum comprises softeners in an amount of about 0 to about 18% by weight of the chewing gum, more typically about 0 to about 12 % by weight of the chewing gum.

In an embodiment of the invention, the chewing gum ingredients comprise active ingredients.

10

In an embodiment of the invention, said active ingredients are selected from the group of: Acetaminophen, Acetylsalicylsyre Buprenorphine Bromhexin Celcoxib Codeine, Diphenhydramin, Diclofenac, Etoricoxib, Ibuprofen, Indometacin, Ketoprofen, Lumiracoxib, Morphine, Naproxen, Oxycodon, Parecoxib, Piroxicam, Pseudoefedrin, Rofecoxib, Tenoxicam, Tramadol, Valdecoxib, Calciumcarbonat, Magaldrate, Disulfiram, Bupropion, Nicotine, Azithromycin, Clarithromycin, Clotrimazole, Erythromycin, Tetracycline, Granisetron, Ondansetron, Prometazin, Tropisetron, Brompheniramine, Ceterizin, Ieco-Ceterizin, Chlorcyclizine, Chlorpheniramin, Chlorpheniramin, Difenhydramine, Doxylamine, Fenofenadin, Guaifenesin, Loratidin, des-Loratidin, Phenyltoloxamine, Promethazin, Pyridamine, Terfenadin, Troxerutin, Methyldopa, Methylphenidate, Benzalcon. Chloride, Benzeth. Chloride, Cetylpyrid. Chloride, Chlorhexidine, Ecabet-sodium, Haloperidol, Allopurinol, Colchicine, Theophylline, Propanolol, Prednisolone, Prednisone, Fluoride, Urea, Miconazole, Actot, Glibenclamide, Glipizide, Metformin, Miglitol, Repaglinide, Rosiglitazone, Apomorfin, Cialis, Sildenafil, Vardenafil, Diphenoxylate, Simethicone, Cimetidine, Famotidine, Ranitidine, Ratinidine, cetirizin, Loratadine, Aspirin, Benzocaine, Dextrometorphan, Ephedrine, Phenylpropanolamine, Pseudoephedrine, Cisapride, Domperidone, Metoclopramide, Acyclovir, Dioctylsulfosucc., Phenolphthalein, Almotriptan, Eletriptan, Ergotamine, Migea, Naratriptan, Rizatriptan, Sumatriptan, Zolmitriptan, Aluminium salts, Calcium salts, Ferro salts, Silver salts, Zinc-salte, Amphotericin B, Chlorhexidine, Miconazole, Triamcinolonacetomid, Melatonine, Phenobarbitol, Caffeine,

30

Benzodiazepiner, Hydroxyzine, Meprobamate, Phenothiazine, Buclizine, Brometazine, Cinnarizine, Cyclizine, Difenhhydramine, Dimenhydrinate, Buflomedil, Amphetamine, Caffeine, Ephedrine, Orlistat, Phenylephedrine, Phenylpropanolamin, Pseudoephedrine, Sibutramin, Ketoconazole, Nitroglycerin, Nystatin, Progesterone, 5 Testosterone, Vitamin B12, Vitamin C, Vitamin A, Vitamin D, Vitamin E, Pilocarpin, Aluminiumaminoacetat, Cimetidine, Esomeprazole, Famotidine, Lansoprazole, Magnesiumoxide, Nizatide and or Ratinidine or derivates and mixtures thereof.

- 10 In an embodiment of the invention, the chewing gum is substantially free of non-biodegradable polymers

In an embodiment of the invention the at least two ore more cyclic esters are selected from the groups of glycolides, lactides, lactones, cyclic carbonates or mixtures 15 thereof.

In an embodiment of the invention the lactone monomers are chosen from the group of ϵ -caprolactone, δ -valerolactone, γ -butyrolactone, and β -propiolactone. It also includes ϵ -caprolactones, δ -valerolactones, γ -butyrolactones, or β -propiolactones that 20 have been substituted with one or more alkyl or aryl substituents at any non-carbonyl carbon atoms along the ring, including compounds in which two substituents are contained on the same carbon atom.

In an embodiment of the invention the carbonate monomer is selected from the group 25 of trimethylene carbonate, 5-alkyl-1,3-dioxan-2-one, 5,5-dialkyl-1,3-dioxan-2-one, or 5-alkyl-5-alkyloxycarbonyl-1,3-dioxan-2-one, ethylene carbonate, 3-ethyl-3-hydroxymethyl, propylene carbonate, trimethylolpropane monocarbonate, 4, 6dimethyl-1, 3-propylene carbonate, 2, 2-dimethyl trimethylene carbonate, and 1, 3-dioxepan-2-one and mixtures thereof.

30

In an embodiment of the invention the cyclic ester polymers and their copolymers resulting from the polymerization of cyclic ester monomers include, but are not

limited to : poly (L-lactide) ; poly (D-lactide) ; poly (D, L-lactide) ; poly (mesolactide) ; poly (glycolide) ; poly (trimethylenecarbonate) ; poly (epsilon-caprolactone) ; poly (L-lactide-co-D, L-lactide) ; poly (L-lactide-co-meso-lactide) ; poly (L-lactide-co-glycolide) ; poly (L-lactide-co-trimethylenecarbonate) ; poly (L-lactide-co-epsilon-caprolactone) ; poly (D, L-lactide-co-meso-lactide) ; poly (D, L-lactide-co-glycolide) ; poly (D, L-lactide-co-trimethylenecarbonate) ; poly (D, L-lactide-co-epsilon-caprolactone) ; poly (meso-lactide-co-glycolide) ; poly (meso-lactide-co-trimethylenecarbonate) ; poly (meso-lactide-co-epsilon-caprolactone) ; poly (glycolide-cotrimethylenecarbonate) ; poly (glycolide-co-epsilon-caprolactone).

In an embodiment of the invention the chewing gum comprises filler.

15 A chewing gum base formulation may, if desired, include one or more fillers/texturisers including as examples, magnesium and calcium carbonate, sodium sulphate, ground limestone, silicate compounds such as magnesium and aluminium silicate, kaolin and clay, aluminium oxide, silicium oxide, talc, titanium oxide, mono-, di- and tri-calcium phosphates, cellulose polymers, such as wood, and
20 combinations thereof.

In an embodiment of the invention the chewing gum comprises filler in an amount of about 0 to about 50% by weight of the chewing gum, more typically about 10 to about 40 % by weight of the chewing gum.

25 In an embodiment of the invention the chewing gum comprises at least one coloring agent.

According to an embodiment of the invention, the chewing gum may comprise color
30 agents and whiteners such as FD&C-type dyes and lakes, fruit and vegetable extracts, titanium dioxide and combinations thereof. Further useful chewing gum

base components include antioxidants, e.g. butylated hydroxytoluene (BHT), butyl hydroxyanisol (BHA), propylgallate and tocopherols, and preservatives.

In an embodiment of the invention the chewing gum is coated with an outer coating.

5

In an embodiment of the invention the outer coating is a hard coating.

In an embodiment of the invention the hard coating is a coating selected from the group consisting of a sugar coating and a sugarless coating and a combination thereof.

10

In an embodiment of the invention the hard coating comprises 50 to 100% by weight of a polyol selected from the group consisting of sorbitol, maltitol, mannitol, xylitol, erythritol, lactitol and isomalt.

15

In an embodiment of the invention the outer coating is an edible film comprising at least one component selected from the group consisting of an edible film-forming agent and a wax.

20 In an embodiment of the invention the film-forming agent is selected from the group consisting of a cellulose derivative, a modified starch, a dextrin, gelatine, shellac, gum arabic, zein, a vegetable gum, a synthetic polymer and any combination thereof.

In an embodiment of the invention the outer coating comprises at least one additive component selected from the group consisting of a binding agent, a moisture absorbing component, a film forming agent, a dispersing agent, an antisticking component, a bulking agent, a flavouring agent, a colouring agent, a pharmaceutically or cosmetically active component, a lipid component, a wax component, a sugar, an acid and an agent capable of accelerating the after-chewing degradation of the degradable polymer.

30

In an embodiment of the invention the outer coating is a soft coating.

In an embodiment of the invention the soft coating comprises a sugar free coating agent.

- 5 In an embodiment of the invention the chewing gum comprises conventional chewing gum polymers or resins.

In an embodiment of the invention the at least one biodegradable polymer comprises at least 5% of the chewing gum polymers.

10

In an embodiment of the invention all the biodegradable polymers comprised in the chewing gum comprises at least 25%, preferably at least 50% of the chewing gum polymers.

- 15 In an embodiment of the invention the biodegradable polymers comprised in the chewing gum comprises at least 80%, preferably at least 90% of the chewing gum polymers.

- In an embodiment of the invention the chewing gum comprises
20 said at least one biodegradable polyester copolymer forming a plasticizer of the chewing gum and
at least one non-biodegradable conventional elastomer

- According to the invention, a biodegradable polymer according to the invention may
25 form a substitute of a conventional natural or synthetic resin.

- In an embodiment of the invention the chewing gum comprises
the at least one biodegradable polyester copolymer forming an elastomer of the
chewing gum and at least one non-biodegradable conventional natural or synthetic
30 resin.

According to the invention, a biodegradable polymer according to the invention may form a substitute of a conventional low or high molecular weight elastomer.

In an embodiment of the invention said chewing gum comprises

5

at least one biodegradable elastomer in the amount of about 0.5 to about 70% wt of the chewing gum,

at least one biodegradable plasticizer in the amount of about 0.5 to about 70% wt of
10 the chewing gum and

at least one chewing gum ingredient chosen from the groups of softeners, sweeteners, flavoring agents, active ingredients and fillers in the amount of about 2 to about 80% wt of the chewing gum.

15

The figures

The invention will now be described with reference to the drawings of which

20 fig. 1 illustrates a transcarbocation reaction during stannous octoate-catalyzed ring-opening polymerization,

fig. 2 to 5 and 10 to 12 illustrate different measured texture properties of the obtained biodegradable chewing gum polymer and where

25

fig. 6 to 9 illustrate the measured LVR properties of the obtained polymers when incorporated in chewing gum at the chewing times 15, 30, 60 and 120 seconds, respectively.

30 fig. 13 to 16 illustrate release properties of the obtained polymers when incorporated in chewing gum.

Detailed description

The following examples of the invention are non-limiting and only provided for the purpose of explaining the invention.

Unless otherwise indicated, as used herein, the term "molecular weight" means number average molecular weight (M_n).

- 10 It has surprisingly been found that biodegradable elastomers, suitable for the formulation of chewing gum base, can be made by metal-catalyzed ring-opening polymerization using a combination of an initiator comprising a trifunctional or higher polyol and a mixture of cyclic monomers including lactones and at least one cyclic carbonate monomer. These polymers derive their excellent elastomeric
- 15 properties from the fact that they are non-crystallizable polymers with a glass transition temperature below room temperature, and they are hyperbranched or lightly crosslinked materials, which characteristic imparts excellent elasticity and recovery.
- 20 The various monomers are strategically selected to impart specific properties to the polymers of the invention. The requirement of non-crystallizability is achieved through the use of two or more monomers that can enter the polymer chain in an approximately random sequence, thus imparting disorder along the backbone. Crystallization is also hindered by the branch point introduced by the trifunctional or
- 25 higher polyol initiator. The monomer representing the major component of the backbone, which should also possess a very low homopolymer glass transition temperature, is selected from the family of aliphatic lactones, with ϵ -caprolactone being a non-limiting example. The comonomer or comonomers used to impart disorder should also be selected from the family of aliphatic lactones, but must be
- 30 different from the major-component monomer. A representative but non-limiting example of a monomer suitable for use with the major-component monomer is δ -valerolactone.

The critical, and perhaps most surprising discovery of the invention is that the addition of a small proportion of a carbonate monomer, of which 1,3-dioxan-2-one (trimethylene carbonate) is a non-limiting example, provides a means for introducing
5 additional branching and/or crosslinking to the elastomeric polymer during ring-opening polymerization. In fact, the level of cyclic carbonate in the monomer mixture yields precise control over the degree of branching and crosslinking in the final polymer. The mechanism by which the cyclic carbonate monomer imparts crosslinking is based upon the known tendency for metal catalysts, of which stannous
10 octoate is a non-limiting example, to promote transesterification and transcarbonation reactions (intermolecular chain transfer to polymer) during polymerization.

A transcarbonation reaction during stannous octoate-catalyzed ring-opening
15 polymerization of lactone and carbonate monomers is illustrated in the fig. 1.

This mechanism is shown in the figures. Fig. 1 illustrate three-arm star polymer molecules produced from a trifunctional polyol initiator (I) such as trimethylolpropane. The backbone of these polymers is composed of randomly
20 incorporated ϵ -caprolactone and trimethylene carbonate mer units, and the ends of each arm carry either a polymerization-active stannyl ether group as illustrated in (1) or a polymerization-inactive hydroxyl group as illustrated in (2). Transesterification (transcarbonation) involves reaction of the stannyl ether group of one chain with an internal ester (carbonate) linkage of another chain. In (3) a transcarbonation reaction
25 between species illustrated (1) and (2) has been obtained, thereby creating the intermediate (3). The latter can decompose to yield two different products because the carbonate linkage has two different acyl-oxygen bonds that may be broken. The decomposition pathway pictured in the figure illustrated scheme is the one of interest because it yields a new species (4) in which two initiator branch points have become
30 connected. This species represents the very early stages of hyperbranching. As similar reactions take place, more and more branching occurs and the system eventually becomes crosslinked. The degree of crosslinking depends upon the

fractional loading of the cyclic carbonate monomer and the polymerization conversion. The alternate decomposition pathway not pictured does not lead to branching and crosslinking. Also, in the absence of a carbonate monomer, branching and crosslinking do not take place.

5

(5) represents the remaining not-branched copolymer

The trifunctional or higher polyol initiators useful in the present invention include glycerol, trimethylolpropane, pentaerythritol, dipentaerythritol and ethoxylated or propoxylated polyamines. The preferred initiators are trimethylolpropane and pentaerythritol.

10

The monomer representing the major component of the backbone, and the comonomer or comonomers used to impart disorder may be chosen from the same group. This group includes ϵ -caprolactone, δ -valerolactone, γ -butyrolactone, and β -propiolactone. It also includes ϵ -caprolactones, δ -valerolactones, γ -butyrolactones, or β -propiolactones that have been substituted with one or more alkyl or aryl substituents at any non-carbonyl carbon atoms along the ring, including compounds in which two substituents are contained on the same carbon atom. The preferred major component monomer is ϵ -caprolactone. The preferred comonomer is δ -valerolactone.

15

20

The carbonate monomers useful in the present invention include trimethylene carbonate, 5-alkyl-1,3-dioxan-2-one, 5,5-dialkyl-1,3-dioxan-2-one, or 5-alkyl-5-alkyloxycarbonyl-1,3-dioxan-2-one. The preferred carbonate monomer is trimethylene carbonate.

25

In general, the level of crosslinking and the level of hyperbranching would scale approximately the same, that is, if one were high or low, so would the other one be.

30

In general the larger is the ratio carbonate monomer/initiator, the higher the level of hyperbranching and crosslinking.

During polymerization at high temperature, a small fraction of the polymer chains contains catalyst as a part of their structure. The catalyst is transferred from chain to chain in a rapid chemical equilibrium. After polymerization, upon cooling and after
5 polymer workup, the catalyst is believed to not be part of the polymer structure.

EXAMPLE 1

10 Preparation of resin

A resin sample was produced using a cylindrical glass, jacketed 10 L pilot reactor equipped with glass stir shaft and Teflon stir blades and bottom outlet. Heating of the reactor contents was accomplished by circulation of silicone oil, thermostated to
15 130°C, through the outer jacket. D,L-lactide (4.877 kg, 33.84 mol) was charged to the reactor and melted by heating to 140°C for 6 h. After the D,L-lactide was completely molten, the temperature was reduced to 130°C, and stannous octoate (1.79 g, 4.42×10^{-3} mol), 1,2-propylene glycol (79.87 g, 1.050 mol), and ϵ -caprolactone (290.76 g, 2.547 mol) were charged to the reactor. After the mixture
20 became homogeneous, stirring was continued for 24 h at 130°C. At the end of this time, the bottom outlet was opened, and molten polymer was allowed to drain into a Teflon-lined paint can.

Characterization of the product indicated $M_n = 5,700$ g/mol and $M_w = 7,100$ g/mol
25 (gel permeation chromatography with online MALLS detector) and $T_g = 30.7^\circ\text{C}$ (DSC, heating rate $10^\circ\text{C}/\text{min}$).

EXAMPLE 2

30 Preparation of LMWE elastomer

A 515 g LMWE sample was synthesized within a dry N₂ glove box, as follows. Into a 500 mL resin kettle equipped with overhead mechanical stirrer, 0.73 g 1,2-propane diol (3.3 mL of a 22.0% (w/v) solution in methylene chloride), and 0.152 g Sn(Oct)₂ (3.56 mL of a 4.27% (w/v) solution in methylene chloride) were charged under dry N₂ gas purge. The methylene chloride was allowed to evaporate under the N₂ purge for 15 min. Then ϵ -caprolactone (300 g, 2.63 mol) and δ -valerolactone (215 g, 2.15 mol) were added. The resin kettle was submerged in a 130°C constant temperature oil bath and stirred for 14 h. Subsequently the kettle was removed from the oil bath and allowed to cool at room temperature. The solid, elastic product was removed in small pieces using a knife, and placed into a plastic container.

Characterization of the product indicated $M_n = 59,900$ g/mol and $M_w = 74,200$ g/mol (gel permeation chromatography with online MALLS detector) and $T_g = -70^\circ\text{C}$ (DSC, heating rate 10°C/min).

15

EXAMPLE 3

Preparation of HMWE made with difunctional initiator

20

A HMWE sample was synthesized within a dry N₂ glove box, as follows. Into a 500 mL resin kettle equipped with overhead mechanical stirrer, 0.51 g 1,2-propane diol (2.3 mL of a 22.0 % (w/v) solution in MeCl₂), and 0.15 g Sn(Oct)₂ (2.6 mL of a 5.83 % (w/v) solution of in MeCl₂) were charged under dry N₂ gas purge. The MeCl₂ was allowed to evaporate under the N₂ purge for 15 min. Then ϵ -caprolactone (274 g, 2.40 mol), TMC (49 g, 0.48 mol), and δ -valerolactone (192 g, 1.92 mol) were added. The resin kettle was submerged in a 130°C constant-temperature oil bath and stirred for 14 h. Subsequently the kettle was removed from the oil bath and allowed to cool to room temperature. The solid, elastic product was removed in small pieces using a knife, and placed into a plastic container.

30

Characterization of the product indicated $M_n = 72,400$ g/mol and $M_w = 103,300$ g/mol (gel permeation chromatography with online MALLS detector) and $T_g = -66^\circ\text{C}$ (DSC, heating rate $10^\circ\text{C}/\text{min}$).

5

EXAMPLE 4

Preparation of HMWE made with 4-arms starshaped initiator

- 10 A HMWE sample according to the invention was synthesized in a dry N_2 glove box, as follows. Into a 500 mL resin kettle equipped with overhead mechanical stirrer was charged 0.037 g $\text{Sn}(\text{Oct})_2$ (3.4 ml of a 1.10% (w/v) solution in methylene chloride) under dry N_2 gas purge. The methylene chloride was allowed to evaporate under the N_2 purge for 15 min. Then, pentaerythritol (0.210 g, 1.54×10^{-3} mol), ϵ -
- 15 caprolactone (79.0g, 0.692 mol), TMC(8.0 g, 0.078 mol) and δ -valerolactone (38.0 g, 0.380 mol) were added. The resin kettle was submerged in a 130°C constant temperature oil bath and stirred for 14 h. Subsequently the kettle was removed from the oil bath and allowed to cool at room temperature. The solid, elastic product was removed in small pieces using a knife, and placed into a plastic container.
- 20 Characterization of the product indicated $M_n = 64,600$ g/mol and $M_w = 165,200$ g/mol (gel permeation chromatography with online MALLS detector) and $T_g = -66^\circ\text{C}$ (DSC, heating rate $10^\circ\text{C}/\text{min}$).

25

EXAMPLE 5

Preparation of gumbases

All the gumbases are prepared with following basic formulation:

30

IngredientsPercent by weight

Elastomer HMWE	20
Elastomer LMWE	40
Resin	40

No	Type	Elastomer HMWE	Elastomer LMWE	Resin
101	Standard	Polyisobutylene Mn =73.000	Polyisobutylene Mn =30.000	Polyvinylacetate Mn =5000
102	2-arms initiator	Elastomer polymer from example 3	Elastomer polymer from example 2	Resin polymer from example 1
103	4-arms initiator	Elastomer polymer from example 4	Elastomer polymer from example 2	Resin polymer from example 1

5 Table 1: Gumbase preparation

The gumbases are prepared as follows:

10 HMWE elastomer is added to a mixing kettle provided with mixing means like e.g. horizontally placed Z-shaped arms. The kettle had been preheated for 15 minutes to a temperature of about 60-80°C. The rubber is broken into small pieces and softened with mechanical action on the kettle.

15 The resin is slowly added to the elastomer until the mixture becomes homogeneous. The remaining resin is then added to the kettle and mixed for 10-20 minutes. The LMWE elastomer is added and mixed for 20-40 minutes until the whole mixture becomes homogeneous.

20 The mixture is then discharged into the pan and allowed to cool to room temperature from the discharged temperature of 60-80°C, or the gumbase mixture is used directly for chewing gum by adding all chewing gum components in an appropriate order under continuous mixing.

25 EXAMPLE 6

Preparation of Chewing gum

All chewing gum formulations are prepared with the following basic formulation

5

Peppermint:

	<u>Ingredients</u>	<u>Percent by weight</u>
10	Gum base	40
	Sorbitol	48.6
	Lycasin	3
	Peppermint oil	1.5
	Menthol crystals	0.5
15	Aspartame	0.2
	Acesulfame	0.2
	Xylitol	6

	Type	Gumbase
1001	std	101
1002	difunctional initiator	102
1003	4-arms starshaped initiator	103

Table 2: Peppermint chewing gum preparation

20

Strawberry:

	<u>Ingredients</u>	<u>Percent by weight</u>
25	Gum base	40
	Sorbitol	46.7
	Lycasin	3
	Lecithin	0.3

	Wild Strawberry oil	2
	Apple acid	0.5
	Citric acid	1.1
	Aspartame	0.3
5	Acesulfame	0.1
	Xylitol	6

	Type	Gumbase
1004	Difunctional initiator	102
1005	4-arms starshaped initiator	103

Table 3: Strawberry chewing gum preparation

10

The chewing gum products are prepared as follows:

The gumbase is added to a mixing kettle provided with mixing means like e.g. horizontally placed Z-shaped arms. The kettle had been preheated for 15 minutes to a temperature of about 60-80°C. Or the chewing gum is one step, immediately after preparation of gumbase in the same mixer where the gum base and kettle have a temperature of about 60-80°C.

Mint formulation:

20

One third portion of the sorbitol is added together with the gum base and mixed for 1-2 minutes. Another one third portion of the sorbitol and lycasin is then added to the kettle and mixed for 2 minutes. The remaining one third portion of sorbitol, peppermint and menthol are added and mixed for 2 minutes. Then aspartame and acesulfame are added to the kettle and mixed for 3 minutes. Xylitol is added and mixed for 3 minutes. The resulting gum mixture is then discharged and e.g. transferred to a pan at temperature of 40-48°C. The gum is then rolled and scored into cores, sticks, balls, cubes, and nay other desired shape, optionally followed by coating and polishing processes prior to packaging.

Strawberry formulation:

One third portion of the sorbitol is added together with the gum base and mixed for
5 1-2 minutes. Another one third portion of the sorbitol, lycasin and lecithin are then
added to the kettle and mixed for 2 minutes. The remaining one third portion of
sorbitol, strawberry and acids are added and mixed for 2 minutes. Then aspartame
and acesulfame are added to the kettle and mixed for 3 minutes. Xylitol is added and
mixed for 3 minutes. The resulting gum mixture is then discharged and e.g.
10 transferred to a pan at temperature of 40-48°C. The gum is then rolled and scored
into cores, sticks, balls, cubes, and any other desired shape, optionally followed by
coating and polishing processes prior to packaging.

15 EXAMPLE 7

An experiment was set up in order to test if the 4-arms starshaped HMWE elastomer
has a closer rheological match, to conventional HMWE elastomer e.g. polyisobutylene
or butylrubber, compared with a HMWE elastomer made with a difunctional
20 initiator.

Accordingly, the following rheological parameters were measured using a
rheometer, type AR1000 from TA Instruments. The oscillation measurement is
performed at a stress within the linear viscoelastic region and a temperature of 130°C
25 with a parallel plate system (d=2.0 cm, hatched). G' , and $\tan \delta$ vs. shear rate.

The results are summarised in fig.2, 3 and as it appears, the elasticity of the
elastomer made with 4-arms star shaped initiator was much closer to the
conventional elastomer than the elastomer with a difunctional initiator. The same
30 appears when looking at storage modulus G' .

EXAMPLE 8

An experiment was set up in order to test gumbases, prepared according to EXAMPLE 5, containing the same elastomers described in EXAMPLE 7.

5

Thus, a standard gum base containing 20% HMWE PIB (sample 101, table 1) was compared with a gum base containing 20 % HMWE elastomer made with difunctional initiator (sample 102, table 1) and a gum base containing 20 % HMWE elastomer made with 4-arms star shaped initiator (sample 103, table 1). Accordingly, the following rheological parameters G' and $\tan \delta$ vs. shear rate at 130°C were measured using the method and rheometer described in the previous example.

10

The results are summarised in fig.4 and 5 and as it appears, the gumbase containing the star-shaped elastomer (103) gives a closer rheological match to the gumbase containing conventional elastomers (101) compared to gumbase containing elastomer made with a diol initiator (102).

15

EXAMPLE 9

20

Chewing profile

An experiment was set up in order to test the corresponding chewing gum samples to the gum bases described in EXAMPLE 8. Prepared as described in EXAMPLE 6.

25

In order to test the chewing profile of the chewing gum samples containing the gum bases with star shaped biodegradable elastomer, difunctional elastomer and std (samples 1003, 1002 and 1001, respectively). The gum centres were chewed in a chewing machine (CF Jansson). The chewing frequency was set to 1 Hz, a pH buffer was used as saliva and the temperature was set at 37°C. The chewing time was set to 15 seconds, 30 seconds, 60 seconds and 120 seconds. After chewing, the chewed cud

30

was measured on a rheometer, described in EXAMPLE 7 as oscillation measurements at a temperature of 37°C.

5 The results from these measurements can be seen on fig. 6, 7, 8 and 9 wherein the storage modulus (G') versus oscillation torque is depicted at different chewing times illustrating the texture changes during chewing.

10 From fig. 6 it can be seen that while the two chewing gum formulations containing elastomers made from difunctional star shaped initiator (1002) and from multi star shaped initiator (1003) are somewhat softer in the initial phase, after 30 seconds, see fig. 7, the standard (1001) is getting closer to the two others and the sample 1003 is now closer to standard compared with 1002.

15 As illustrated in fig 8 the difference between the three samples is similar to the difference illustrated in fig. 7 after 60 seconds. After 120 seconds, see fig. 9, the difference is smaller, and the values measured on sample 1003 are still closest to the standard formulation 1003.

20 The above rheological results are confirming the fact that the elastomer made with 4-arms star shaped initiator has texture properties closer to conventional elastomers as compared to elastomer made with difunctional initiator, also as a function of time.

EXAMPLE 10

25

Sensory texture profile analyses of test chewing gum

30 The three chewing gum samples were tested by serving them to the sensory panellists in tasting booths made in accordance with ISO 8598 standards at room temperature in 40 ml tasteless plastic cups with randomised 3-figure codes. Test samples were evaluated after chewing for 0-½ minutes (initial phase 1), ½-1 minutes (initial phase 2), 1-1½ minutes (intermediate 1), 1½-2 minutes (intermediate 2), 2-2½ minutes

(intermediate 3), 2½-3 minutes (intermediate 4), 4-4½ minutes (end phase 1), 4½-5 minutes (end phase 2), respectively. Between each sample tested, the panellist were allowed a break of 3 minutes. Every test is repeated.

- 5 The following texture parameters were assessed: softness, toughness and elasticity. For each of these parameters, the panellists were required to provide their assessments according to an arbitrary scale of 0-15. The data obtained were processed using a FIZZ computer program (French Bio System) and the results were transformed to sensory profile diagrams as shown in figure 10-12. The major
10 differences between test chewing gums in all phases were the following:

The chewing gum containing initiator made elastomers (1002, 1003) showed a higher softness compared with standard (confirming the rheological results in the
15 above EXAMPLE 9). When comparing the chewing gum containing initiator made polymers 1002 and 1003, the softness of 1003 (star-shaped) is closer to standard except for the initial phases.

Fig 11 showed a higher toughness of the chewing gum containing elastomer made
20 with 4-arms star shaped initiator (1003) compared with difunctional initiator made elastomer (1002) except for the initials phases. The toughness of 1003 is closer to standard compared with 1002.

The elastisity of 4-arms star shaped elastomer is expected to be higher due to the
25 branching, which is confirmed by fig. 12. Where 1003 was found higher in elasticity and closer to the standard compared with 1002 (made with difunctional initiator) in about 70 % of the time tested.

30 **EXAMPLE 11**

Sensory flavour profile analyses of test chewing gum

The three chewing gum samples were tested using the sensory method described in the above EXAMPLE 10.

Test samples were evaluated after chewing for 0-1 minutes (initial phase 1), 1-2
5 minutes (intermediate phase 1), 2-3 minutes (intermediate phase 2), 3-4 minutes
(intermediate 3), 4-5 minutes (end phase 1), respectively.

The following flavour parameters were assessed: sweetness, flavour intensity and
cooling. For each of these parameters, the panellists were required to provide their
10 assessments according to an arbitrary scale of 0-15. The data obtained were
processed using a FIZZ computer program (French Bio System) and the results were
transformed to sensory profile diagrams as shown in figure 13-15.

The major differences between the chewing gums in all phases were the
15 following:

The chewing gum containing elastomer made with 4-arms star shaped initiator 1003
showed higher sweetness release for the initial phase (fig. 13). Cooling and overall
flavour intensity were found higher in release compared to the chewing gum
20 formulation containing HMWE elastomer made with a difunctional initiator 1002
(fig. 14 and 15).

It can therefore be concluded that the use of a 4-arms star shaped initiator is superior
with regard to essential flavour characteristics.
25

EXAMPLE 12

Sensory time intensity analysis of test chewing gum

30

Two strawberry chewing gum samples were tested by serving them to the sensory panellists in tasting booths made in accordance with ISO 8598 standards at room temperature in 40 ml tasteless plastic cups with randomised 3-figure codes.

5 Samples were tested during 3 minutes and evaluated every 10 seconds. Between each sample tested, the panellist were allowed a break of 3 minutes. Every test is repeated. The FIZZ (French Bio System) is used to collect and calculate data and the results were transformed to sensory time intensity diagram as shown in figure 17.

10 The flavour intensity of strawberry flavoured chewing gum containing elastomer made with 4-arms star shaped initiator 1005 has an higher overall flavours intensity compared with chewing gum formulation containing HMWE elastomer made with a difunctional initiator 1004 (fig. 16).

Patent Claims

1. Degradable chewing gum polymer,
said degradable polymer being a polymer polymerized from
5 at least one trifunctional or higher functional initiator
at least two different monomers forming the backbone of the polymer and
10 at least one monomer selected from the group of carbonate monomers.
2. Degradable chewing gum polymer according to claim 1,
wherein said at least two different monomers are cyclic.
- 15 3. Degradable chewing gum polymer according to claim 1 or 2,
wherein the at least two different monomers forming the backbone of the polymer
comprises at least one backbone monomer and at least one backbone comonomer,
- 20 4. Degradable chewing gum polymer according to any of the claims 1-3,
wherein said at least one backbone comonomer imparts disorder in the backbone
monomer chain.
5. Degradable chewing gum polymer according to any of the claims 1-4,
wherein the at least one backbone comonomer is effective to introduce amorphous
25 regions in the backbone monomer chain.
6. Degradable chewing gum polymer according to any of the claims 1-5,
wherein the at least two different monomers forming the backbone of the polymer
are selected from the group of lactone monomers.
- 30 7. Degradable chewing gum polymer according to any of the claims 1-6,

wherein the lactone monomers are chosen from the group of ϵ -caprolactone, δ -valerolactone, γ -butyrolactone, and β -propiolactone, and also includes ϵ -caprolactones, δ -valerolactones, γ -butyrolactones, or β -propiolactones that have been substituted with one or more alkyl or aryl substituents at any non-carbonyl carbon atoms along the ring, including compounds in which two substituents are contained on the same carbon atom and mixtures thereof.

8. Degradable chewing gum polymer according to any of the claims 1-7,
wherein the at least one backbone monomer comprises ϵ -caprolactone
- 10 9. Degradable chewing gum polymer according to any of the claims 1-8,
wherein the at least one backbone monomer has a Tg below -40°C , preferably less than -50°C .
- 15 10. Degradable chewing gum polymer according to any of the claims 1-9,
wherein the at least one backbone comonomer comprises δ -valerolactone.
11. Degradable chewing gum polymer according to any of the claims 1-10,
wherein said degradable polymer is polymerized by metal catalyzed ring-opening.
- 20 12. Degradable chewing gum polymer according to any of the claims 1-11,
wherein the at least one monomer is selected from the group of carbonate monomers.
13. Degradable chewing gum polymer according to any of the claims 1-12,
25 wherein the at least one monomer selected from the group of carbonate monomers is chosen from the group of trimethylene carbonate, 5-alkyl-1,3-dioxan-2-one, 5,5-dialkyl-1,3-dioxan-2-one, or 5-alkyl-5-alkyloxycarbonyl-1,3-dioxan-2-one, ethylene carbonate, 3-ethyl-3-hydroxymethyl trimethylene carbonate, propylene carbonate, trimethylene carbonate, trimethylolpropane monocarbonate, 4, 6dimethyl-1, 3-propylene carbonate, 2, 2-dimethyl trimethylene carbonate, and 1, 3-dioxepan-2-one
30 and mixtures thereof.

14. Degradable chewing gum polymer according to any of the claims 1-13,
wherein the at least one monomer selected from the group of carbonate monomers
provides a means for introducing additional branching and/or crosslinking to the
elastomeric polymer during ring-opening polymerization.
- 5
15. Degradable chewing gum polymer according to any of the claims 1-14,
wherein said at least one trifunctional or higher functional initiator comprises a
polyol.
- 10
16. Degradable chewing gum polymer according to any of the claims 1-15,
wherein the initiator is selected from the group of glycerol, trimethylolpropane,
pentaerythritol, dipentaerythritol, ethoxylated or propoxylated polyamines and other
molecules with multiple hydroxyl or other reactive groups and other molecules with
multiple hydroxyl or other reactive groups and mixtures thereof.
- 15
17. Degradable chewing gum polymer according to any of the claims 1-16,
wherein the degradable chewing gum polymer is polymerized from:
- about 20 to 80 wt % of the at least one backbone monomer,
- 20 about 19.5 to 79.5 wt % of the at least one backbone comonomer,
about 0.5 to 25 wt % of the at least one monomer selected from the group of
carbonate monomers.
18. Degradable chewing gum polymer according to any of the claims 1-17,
- 25 wherein the degradable chewing gum polymer is moreover polymerized from:
- about 0.01 to 1.0 wt % of the at least one initiator
19. Degradable chewing gum polymer according to any of the claims 1-18,
- 30 wherein the chewing gum properties of the polymer are adjusted by selection of a
suitable order of the multifunctional initiator.

20. Degradable chewing gum polymer according to any of the claims 1-19, wherein the rheological properties of the degradable polymer is controlled by adjusting the functional number of initiator.
- 5 21. Degradable chewing gum polymer according to any of the claims 1-20, wherein the lactone monomers are chosen from the group of ϵ -caprolactone, δ -valerolactone, γ -butyrolactone, and β -propiolactone. It also includes ϵ -caprolactones, δ -valerolactones, γ -butyrolactones, or β -propiolactones that have been substituted with one or more alkyl or aryl substituents at any non-carbonyl carbon atoms along the
- 10 ring, including compounds in which two substituents are contained on the same carbon atom and mixtures thereof.
22. Degradable chewing gum polymer according to any of the claims 1-21, wherein the carbonate monomer is selected from the group of trimethylene
- 15 carbonate, 5-alkyl-1,3-dioxan-2-one, 5,5-dialkyl-1,3-dioxan-2-one, or 5-alkyl-5-alkyloxycarbonyl-1,3-dioxan-2-one, ethylene carbonate, 3-ethyl-3-hydroxymethyl, propylene carbonate, trimethylolpropane monocarbonate, 4, 6dimethyl-1, 3-propylene carbonate, 2, 2-dimethyl trimethylene carbonate, and 1, 3-dioxepan-2-one and mixtures thereof.
- 20 23. Degradable chewing gum polymer according to any of the claims 1-22, wherein the molecular weight of lactone monomers are within the range of 50-16000 g/mol preferably within the range of 100-3000 g/mol
- 25 24. Degradable chewing gum polymer according to any of the claims 1-23, wherein the molecular weight of carbonate monomers are within the range of 50-15000 preferably within the range of 100-2300 g/mol.
25. Chewing gum comprising the degradable polymer according to any of the claims
- 30 1-24.
26. Chewing gum according to claim 25, wherein

said chewing gum ingredients comprise flavoring agents.

27. Chewing gum according to any of claims 25 or 26, wherein
said flavoring agents comprises natural and synthetic flavorings in the form of
5 natural vegetable components, essential oils, essences, extracts, powders, including
acids and other substances capable of affecting the taste profile

28. Chewing gum according to any of claims 25-27, wherein
said chewing gum comprises flavor in an amount of 0.01 to about 30 wt %, said
10 percentage being based on the total weight of the chewing gum

29. Chewing gum according to any of claims 25-28, wherein
said chewing gum comprises flavor in an amount of 0.2 to about 4 wt %, said
percentage being based on the total weight of the chewing gum

15

30. Chewing gum according to any of claims 25- 29, wherein
said flavor comprises water soluble ingredients.

31. Chewing gum according to any of claims 25-30, wherein
20 said water soluble flavor comprises acids.

32. Chewing gum according to any of claims 25-31, wherein
said flavor comprises water insoluble ingredients.

25 33. Chewing gum according to any of claims 25- 32, wherein
said chewing gum ingredients comprising sweeteners.

34. Chewing gum according to any of claims 25-33,
wherein said sweetener comprises bulk sweeteners

30

35. Chewing gum according to any of claims 25-34,

wherein the chewing gum comprises bulk sweeteners in the amount of about 5 to about 95% by weight of the chewing gum, more typically about 20 to about 80% by weight of the chewing gum.

- 5 36. Chewing gum according to any of claims 25-35, wherein said sweetener comprises high intensity sweeteners

37. Chewing gum according to any of claims 25-36, wherein the high intensity sweeteners comprises sucralose, aspartame, salts of acesulfame, alitame, saccharin
10 and its salts, cyclamic acid and its salts, glycyrrhizin, dihydrochalcones, thaumatin, monellin, sterioside, alone or in combination

38. Chewing gum according to any of claims 25-37,
wherein the chewing gum comprises high intensity sweeteners in an amount of about
15 0 to about 1% by weight of the chewing gum, more typically about 0.05 to about 0.5 % by weight of the chewing gum.

39. Chewing gum according to any of claims 25- 38,
wherein the chewing gum comprises at least one softener.

20

40. Chewing gum according to any of claims 25-39,
wherein the at least one softener comprises tallow, hydrogenated tallow,
hydrogenated and partially hydrogenated vegetable oils, cocoa butter, glycerol
monostearate, glycerol triacetate, lecithin, mono-, di- and triglycerides, acetylated
25 monoglycerides, fatty acids - such as stearic, palmitic, oleic and linoleic acids, waxes, PGE and mixtures thereof.

41. Chewing gum according to any of claims 25- 40,
wherein the chewing gum comprises softeners in the amount of about 0 to about 18%
30 by weight of the chewing gum, more typically about 0 to about 12 % by weight of the chewing gum.

42. Chewing gum according to any of claims 25- 41, wherein said chewing gum ingredients comprise active ingredients.
43. Chewing gum according to any of claims 25- 42, said active ingredients being
- 5 selected from the group of: Acetaminophen, Acetylsalicylsyre Buprenorphine
Bromhexin Celcoxib Codeine, Diphenhydramin, Diclofenac, Etoricoxib, Ibuprofen,
Indometacin, Ketoprofen, Lumiracoxib, Morphine, Naproxen, Oxycodon, Parecoxib,
Piroxicam, Pseudoefedrin, Rofecoxib, Tenoxicam, Tramadol, Valdecoxib,
Calciumcarbonat, Magaldrate, Disulfiram, Bupropion, Nicotine, Azithromycin,
- 10 Clarithromycin, Clotrimazole, Erythromycin, Tetracycline, Granisetron,
Ondansetron, Prometazin, Tropisetron, Brompheniramine, Ceterizin, Ieco-Ceterizin,
Chlorcyclizine, Chlorpheniramin, Chlorpheniramin, Difenhydramine, Doxylamine,
Fenofenadin, Guaifenesin, Loratidin, des-Loratidin, Phenyltoloxamine, Promethazin,
Pyridamine, Terfenadin, Troxerutin, Methyldopa, Methylphenidate, Benzalcon.
- 15 Chloride, Benzeth. Chloride, Cetylpyrid. Chloride, Chlorhexidine, Ecabet-sodium,
Haloperidol, Allopurinol, Colchicine, Theophylline, Propanolol, Prednisolone,
Prednisone, Fluoride, Urea, Miconazole, Actot, Glibenclamide, Glipizide,
Metformin, Miglitol, Repaglinide, Rosiglitazone, Apomorfin, Cialis, Sildenafil,
Vardenafil, Diphenoxylate, Simethicone, Cimetidine, Famotidine, Ranitidine,
- 20 Ratinidine, cetirizin, Loratadine, Aspirin, Benzocaine, Dextrometorphan, Ephedrine,
Phenylpropanolamine, Pseudoephedrine, Cisapride, Domperidone, Metoclopramide,
Acyclovir, Dioctylsulfosucc., Phenolphthalein, Almotriptan, Eletriptan, Ergotamine,
Migea, Naratriptan, Rizatriptan, Sumatriptan, Zolmitriptan, Aluminium salts,
Calcium salts, Ferro salts, Silver salts, Zinc-salte, Amphotericin B, Chlorhexidine,
- 25 Miconazole, Triamcinolonacetonid, Melatonine, Phenobarbitol, Caffeine,
Benzodiazepiner, Hydroxyzine, Meprobamate, Phenothiazine, Buclizine,
Brometazine, Cinnarizine, Cyclizine, Difenhydramine, Dimenhydrinate, Buflomedil,
Amphetamine, Caffeine, Ephedrine, Orlistat, Phenylephedrine, Phenylpropanolamin,
Pseudoephedrine, Sibutramin, Ketoconazole, Nitroglycerin, Nystatin, Progesterone,
- 30 Testosterone, Vitamin B12, Vitamin C, Vitamin A, Vitamin D, Vitamin E,
Pilocarpin, Aluminiumaminoacetat, Cimetidine, Esomeprazole, Famotidine,

Lansoprazole, Magnesiumoxide, Nizatide and/or Ratinidine or derivates and mixtures thereof.

44. Chewing gum according to any of claims 25-43, wherein the chewing gum is
5 substantially free of non-biodegradable polymers
45. Chewing gum according to any of claims 25-44,
wherein the chewing gum comprises filler.
- 10 46. Chewing gum according to any of claims 25- 45,
wherein the chewing gum comprises filler in an amount of about 0 to about 50% by
weight of the chewing gum, more typically about 10 to about 40 % by weight of the
chewing gum.
- 15 47. Chewing gum according to any of claims 25-46,
wherein the chewing gum comprises at least one coloring agent..
48. Chewing gum according to any of claims 25-47, where the chewing gum is
coated with an outer coating.
- 20 49. Chewing gum according to any of claims 25-48, wherein the outer coating is a
hard coating.
50. Chewing gum according to any of claims 25-49, wherein the hard coating is a
25 coating selected from the group consisting of a sugar coating and a sugarless coating
and a combination thereof.
51. Chewing gum according to any of claims 25-50, wherein the hard coating com-
prises 50 to 100% by weight of a polyol selected from the group consisting of
30 sorbitol, maltitol, mannitol, xylitol, erythritol, lactitol and isomalt.

52. Chewing gum according to any of claims 25-51, wherein the outer coating is an edible film comprising at least one component selected from the group consisting of an edible film-forming agent and a wax.
- 5 53. Chewing gum according to any of claims 25-52, wherein the film-forming agent is selected from the group consisting of a cellulose derivative, a modified starch, a dextrin, gelatine, shellac, gum arabic, zein, a vegetable gum, a synthetic polymer and any combination thereof.
- 10 54. Chewing gum according to any of claims 25-53, wherein the outer coating comprises at least one additive component selected from the group consisting of a binding agent, a moisture absorbing component, a film forming agent, a dispersing agent, an antisticking component, a bulking agent, a flavouring agent, a colouring agent, a pharmaceutically or cosmetically active component, a lipid component, a wax component, a sugar, an acid and an agent capable of accelerating the after-
15 chewing degradation of the degradable polymer.
55. Chewing gum according to any of claims 25-54, wherein the outer coating is a soft coating.
- 20 56. Chewing gum according to any of claims 25-55, wherein the soft coating comprises a sugar free coating agent.
57. Chewing gum according to any of claims 25-56,
25 wherein said chewing gum comprises conventional chewing gum polymers or resins.
58. Chewing gum according to any of claims 25-57,
wherein the at least one biodegradable polymer comprises at least 5% of the chewing gum polymers.
- 30 59. Chewing gum according to any of claims 25-58,

wherein all the biodegradable polymers comprised in the chewing gum comprises at least 25%, preferably at least 50% of the chewing gum polymers.

60. Chewing gum according to any of claims 25-59,
5 wherein all the biodegradable polymers comprised in the chewing gum comprises at least 80%, preferably at least 90% of the chewing gum polymers.

61. Chewing gum according to any of claims 25-60,
wherein said chewing gum comprises
10 - said at least one biodegradable polyester copolymer forming a plasticizer of the chewing gum and
- at least one non-biodegradable conventional elastomer..

62. Chewing gum according to any of claims 25-61,
15 wherein said chewing gum comprises
- said at least one biodegradable polyester copolymer forming an elastomer of the chewing gum and
- at least one non-biodegradable conventional natural or synthetic resin.

- 20 63. Chewing gum according to any of the claims 25-62, wherein said chewing gum comprises

at least one biodegradable elastomer in the amount of about 0.5 to about 70% wt of the chewing gum,

- 25 at least one biodegradable plasticizer in the amount of about 0.5 to about 70% wt of the chewing gum and

- at least one chewing gum ingredient chosen from the groups of softeners, sweeteners,
30 flavoring agents, active ingredients and fillers in the amount of about 2 to about 80% wt of the chewing gum.

64. Gum base comprising at least one degradable chewing gum polymer according to any of claims 1-24.

1/16

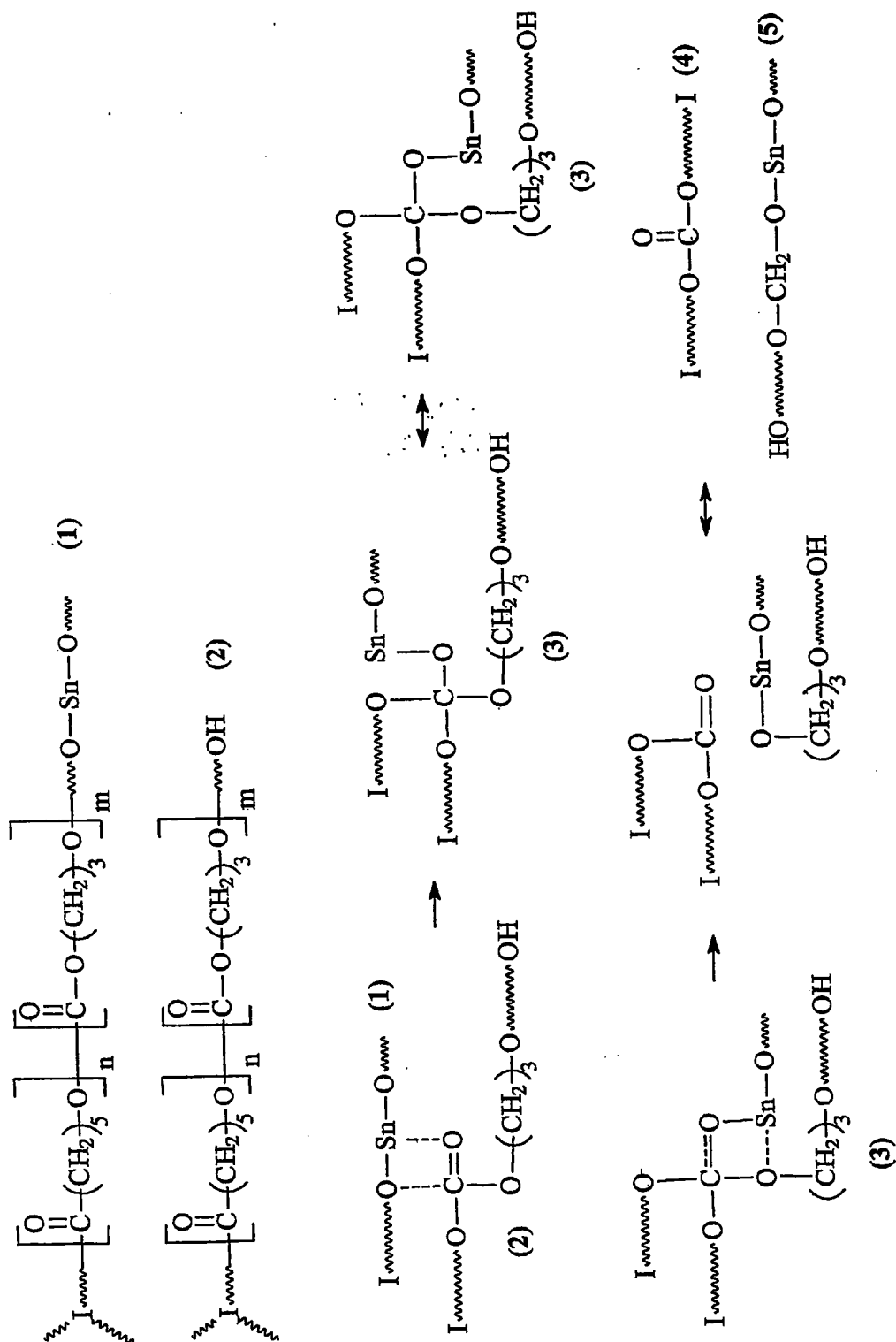


Fig. 1

2/16

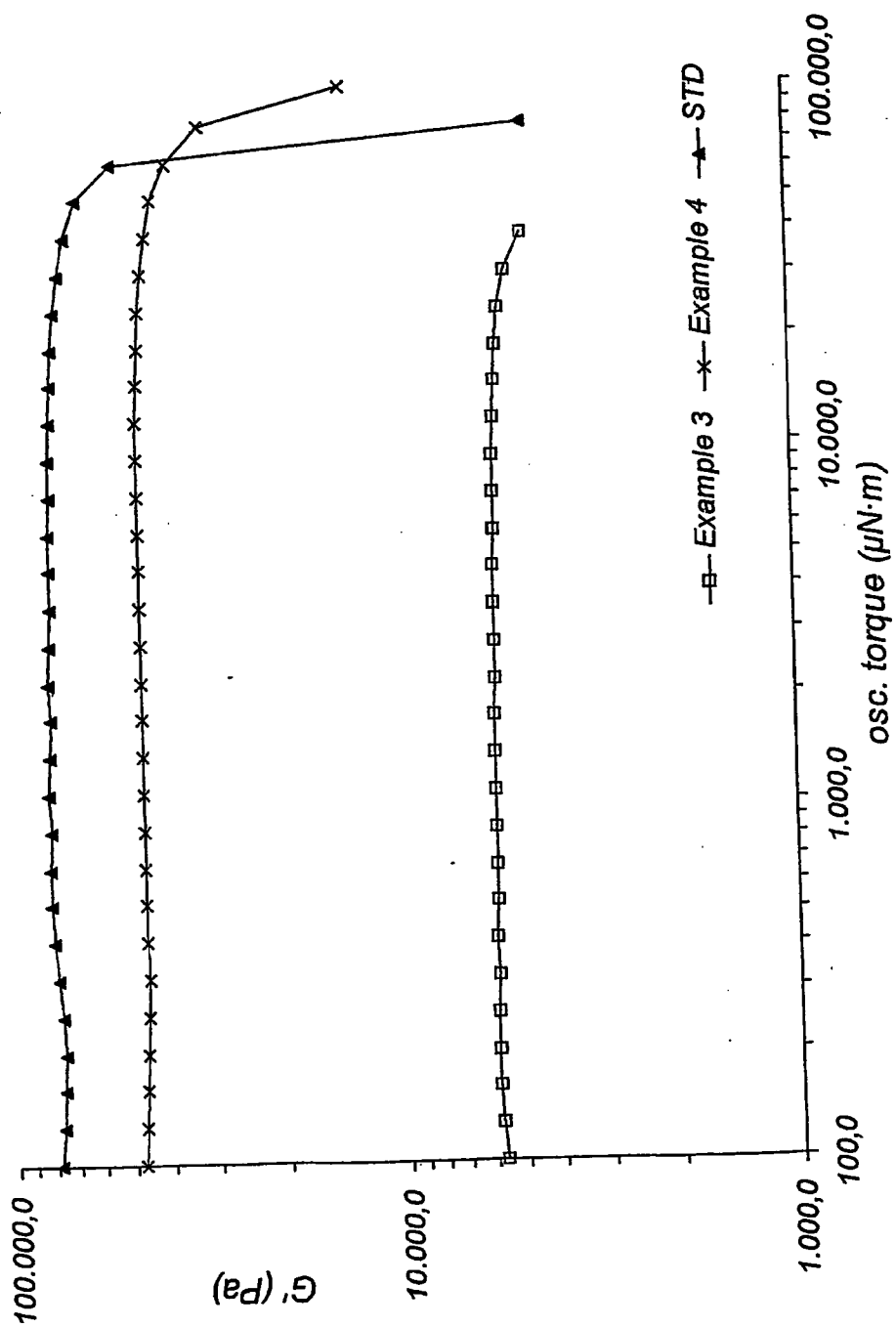


Fig. 2

3/16

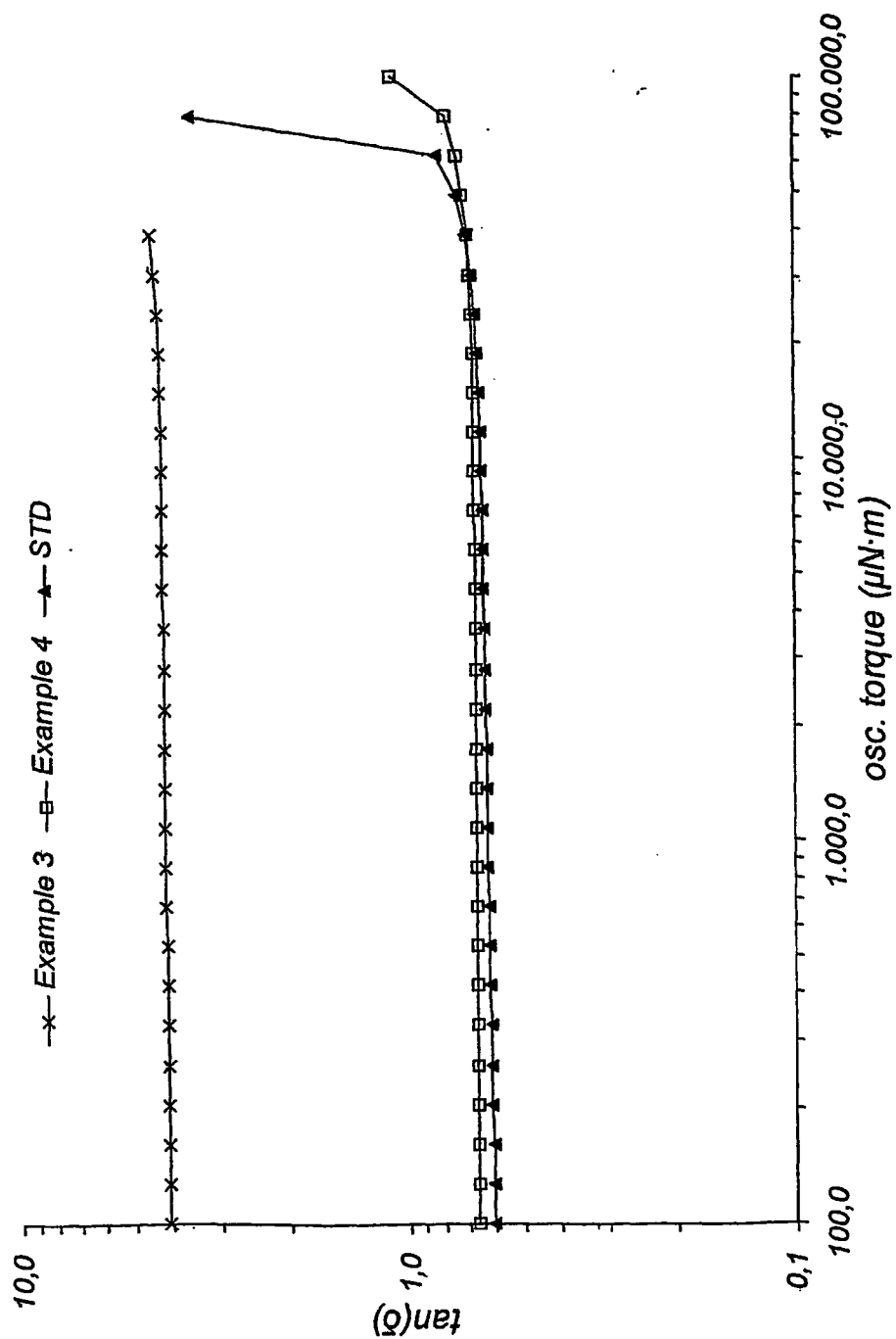


Fig. 3

4/16

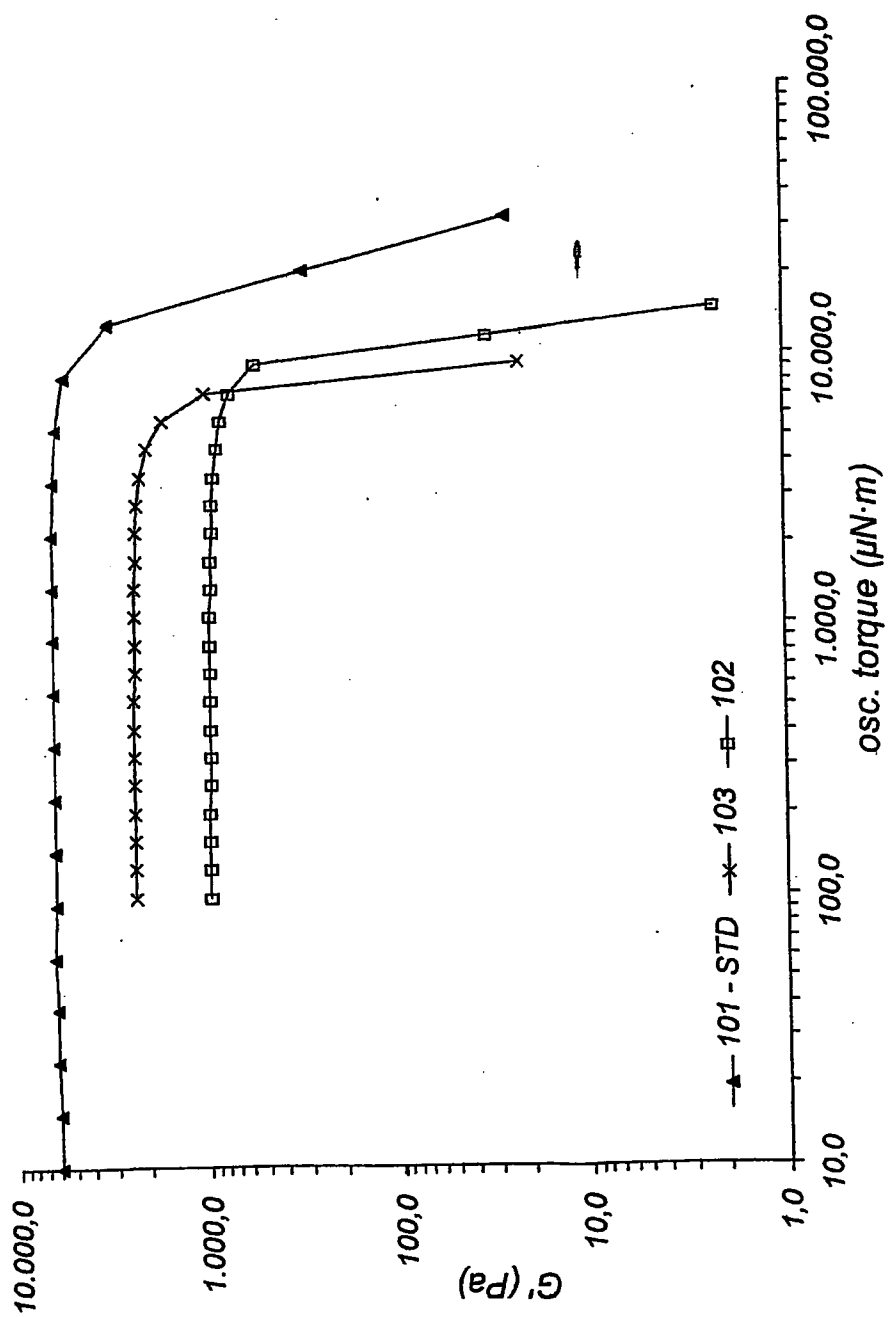


Fig. 4

5/16

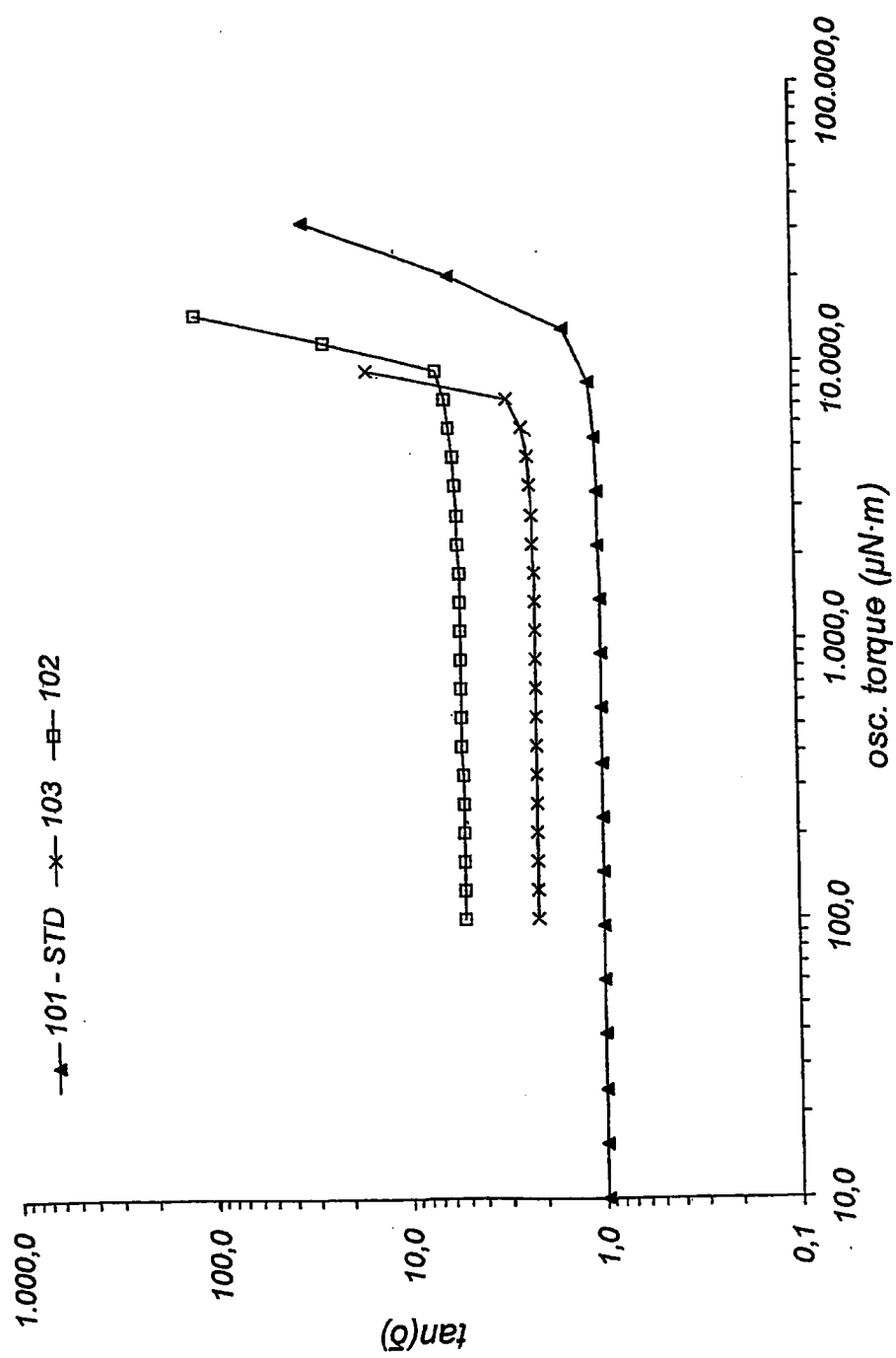


Fig. 5

6/16

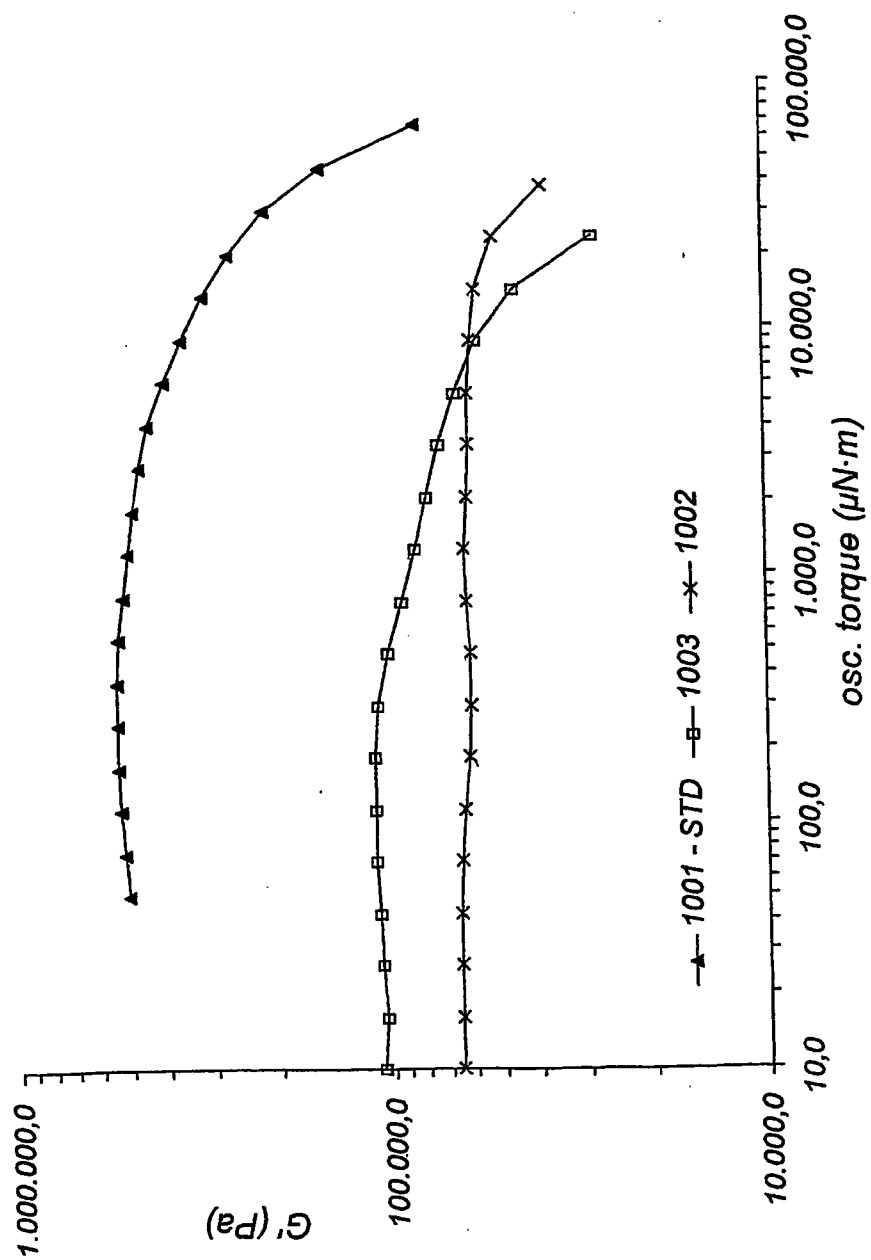


Fig. 6

7/16

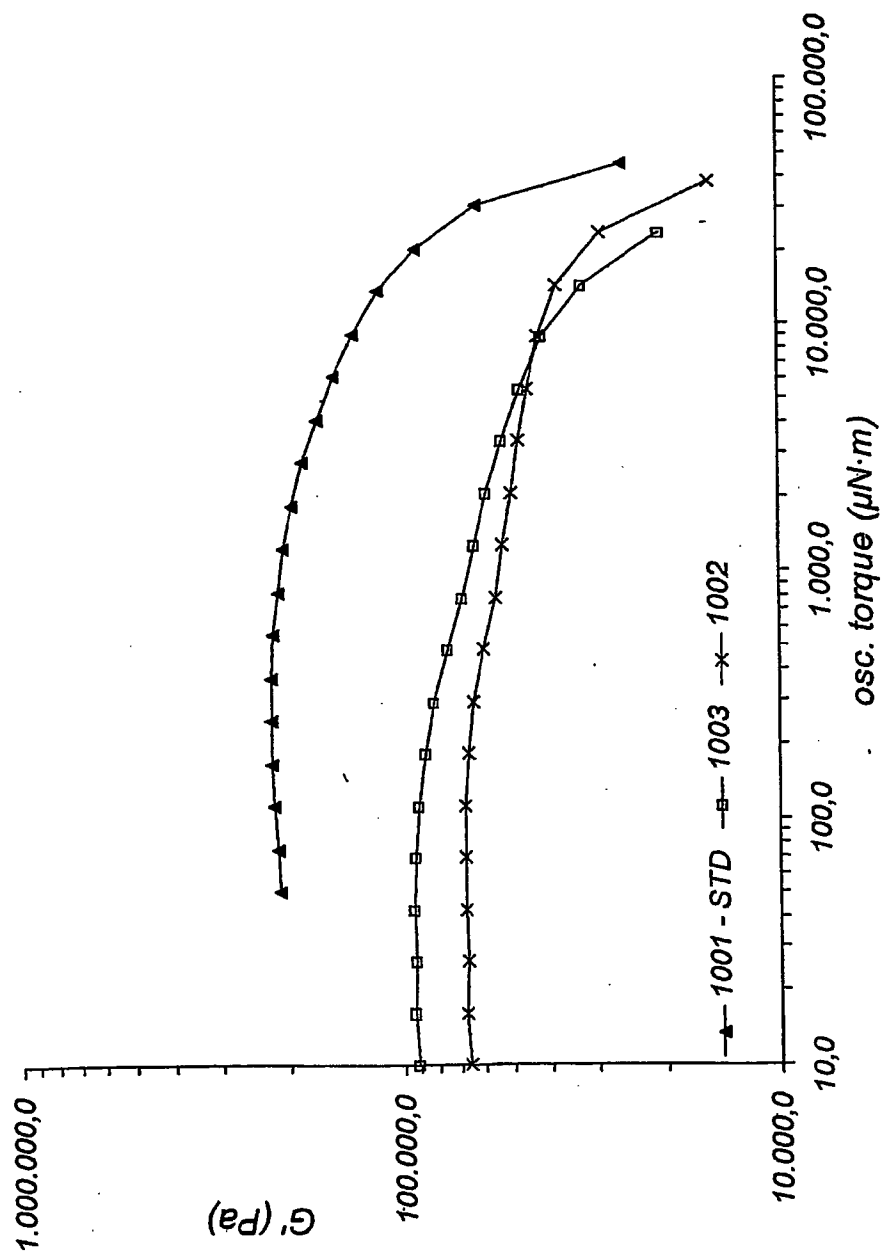


Fig. 7

8/16

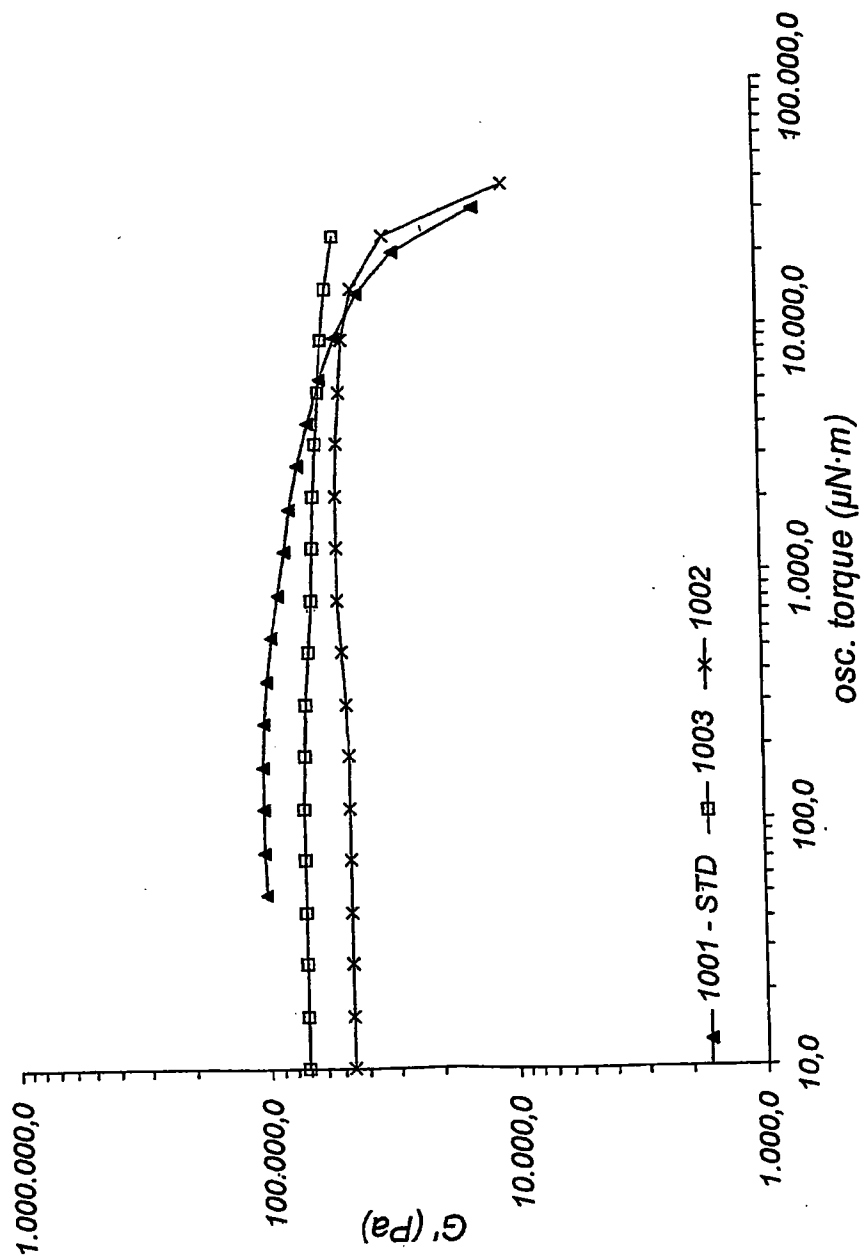


Fig. 8

9/16

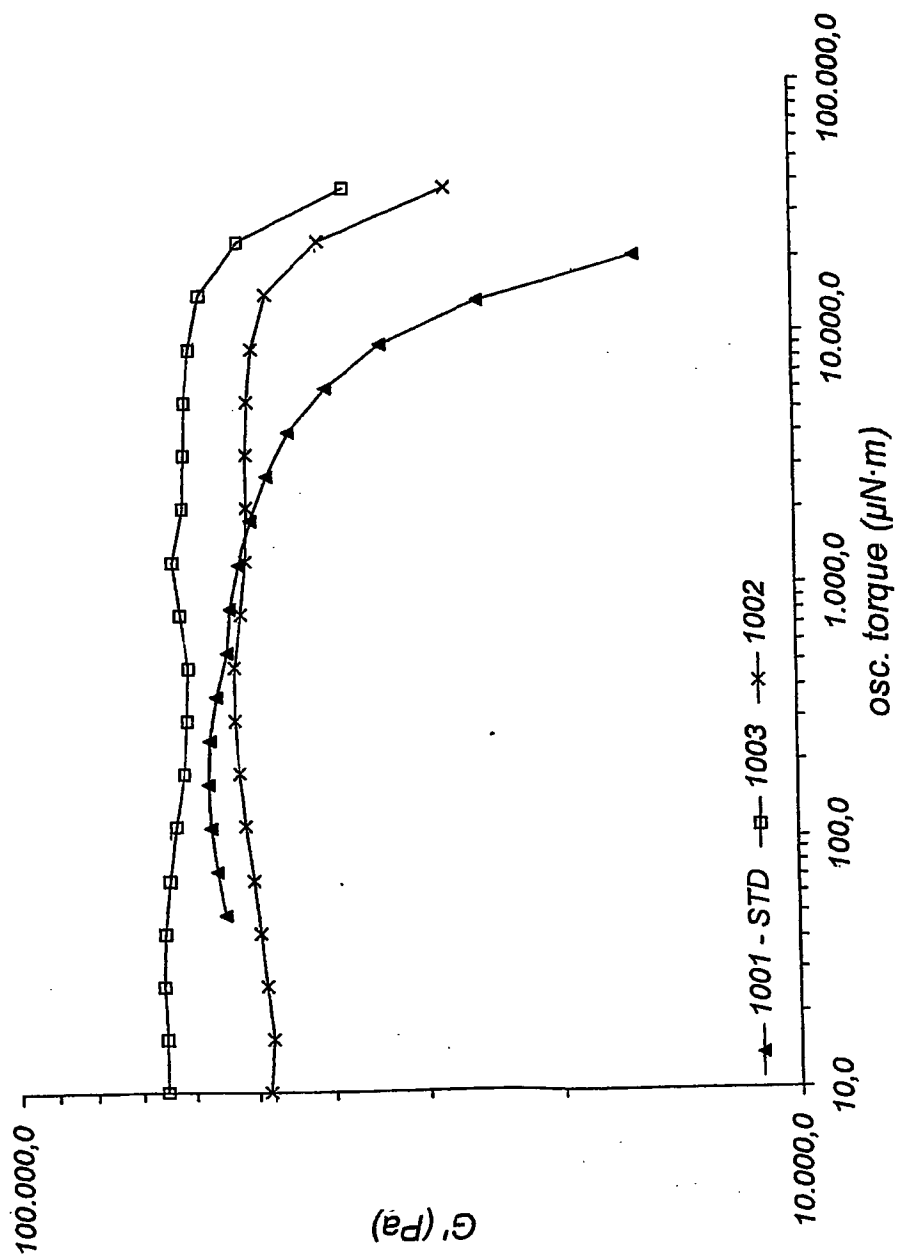


Fig. 9

10 / 16

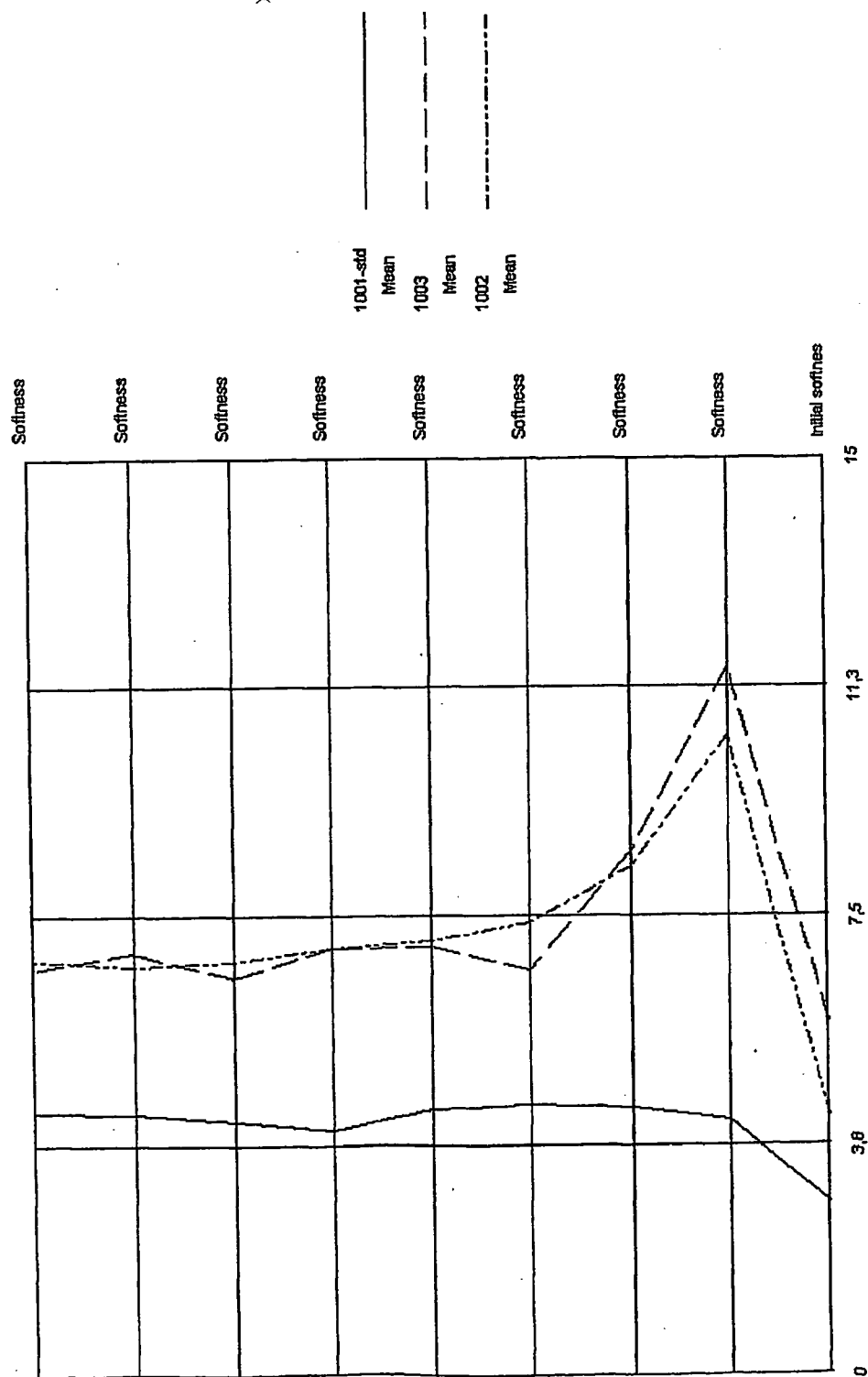


Fig. 10

11 / 16

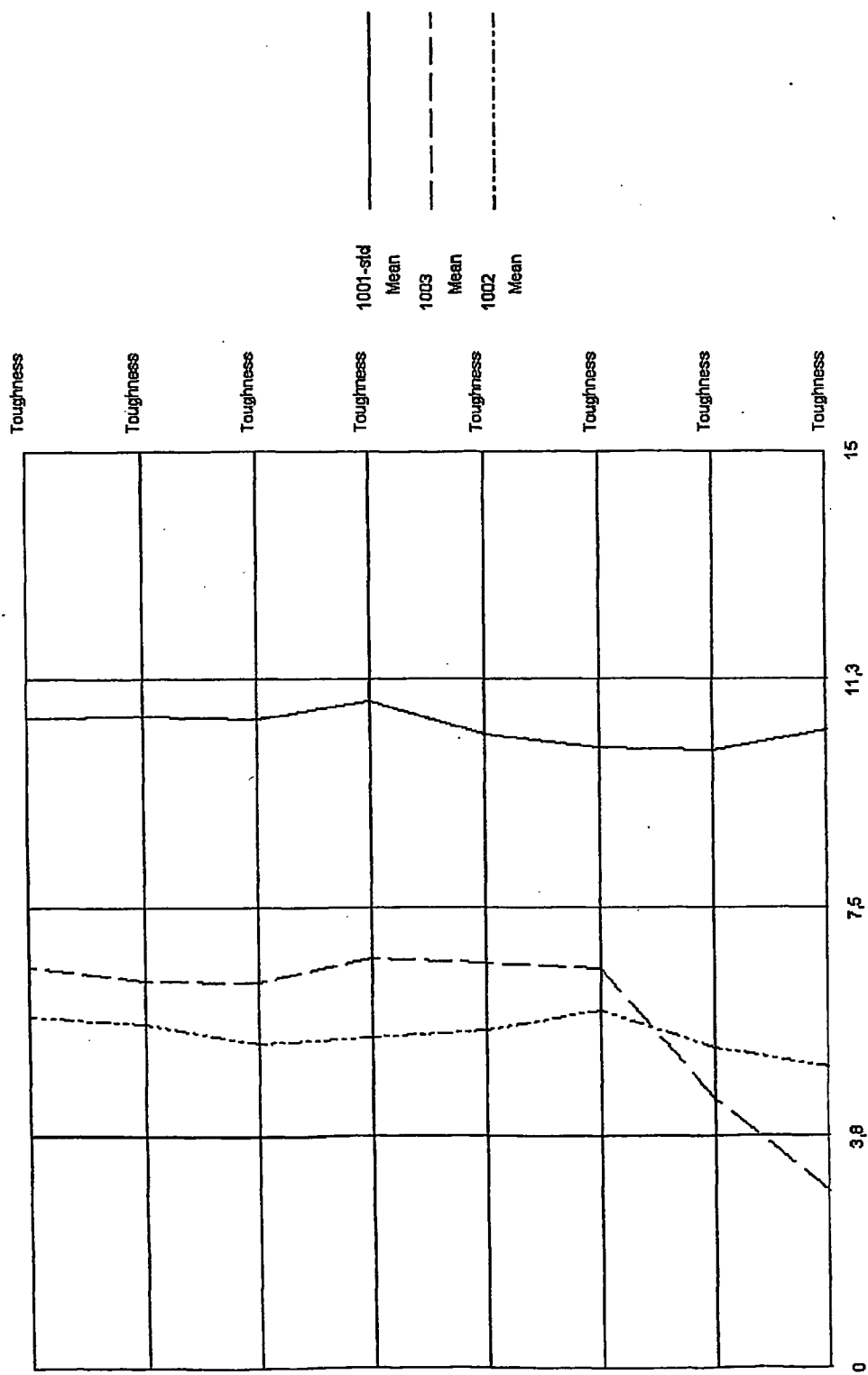


Fig. 11

12 / 16

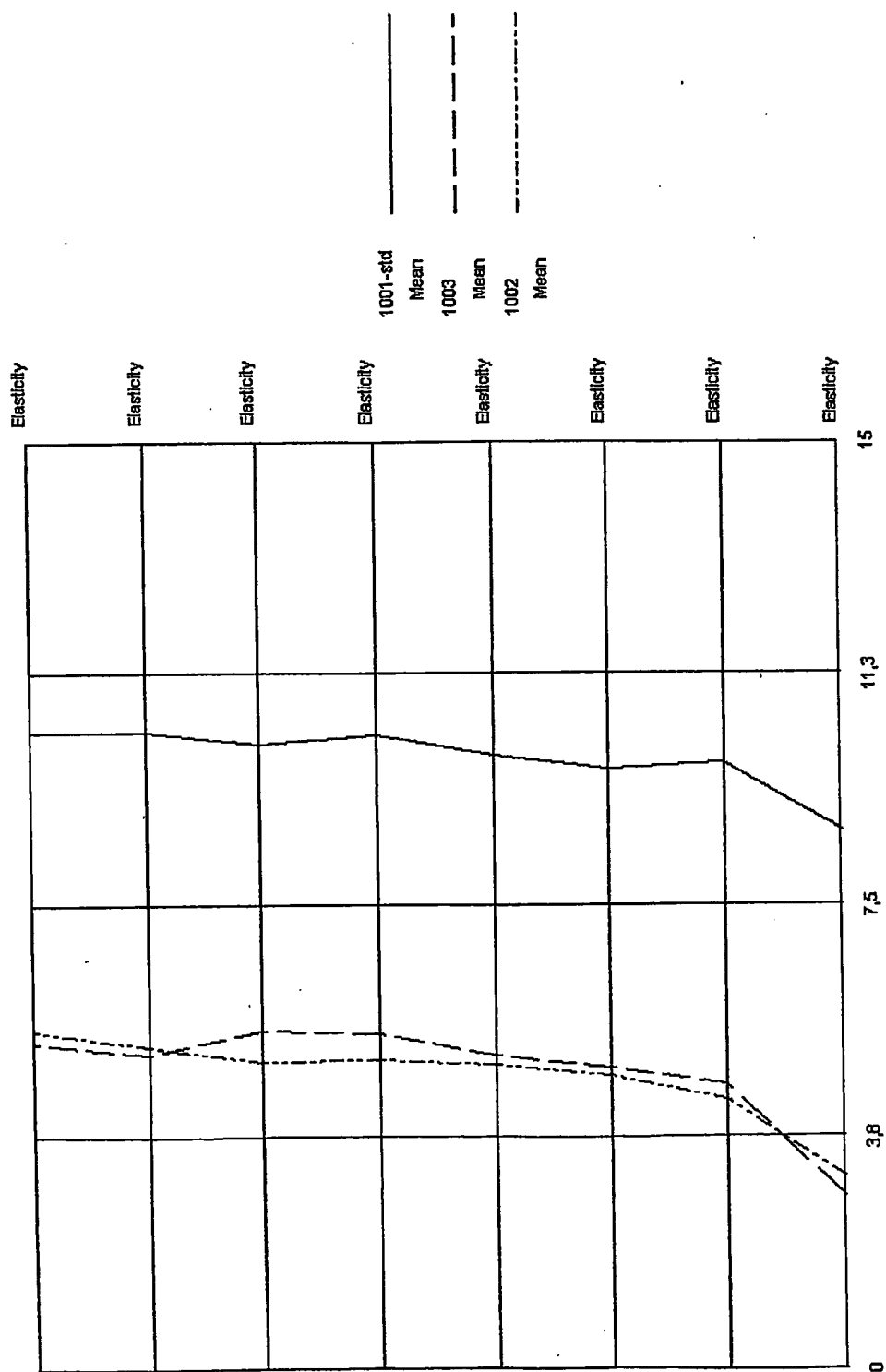


Fig. 12

13 / 16

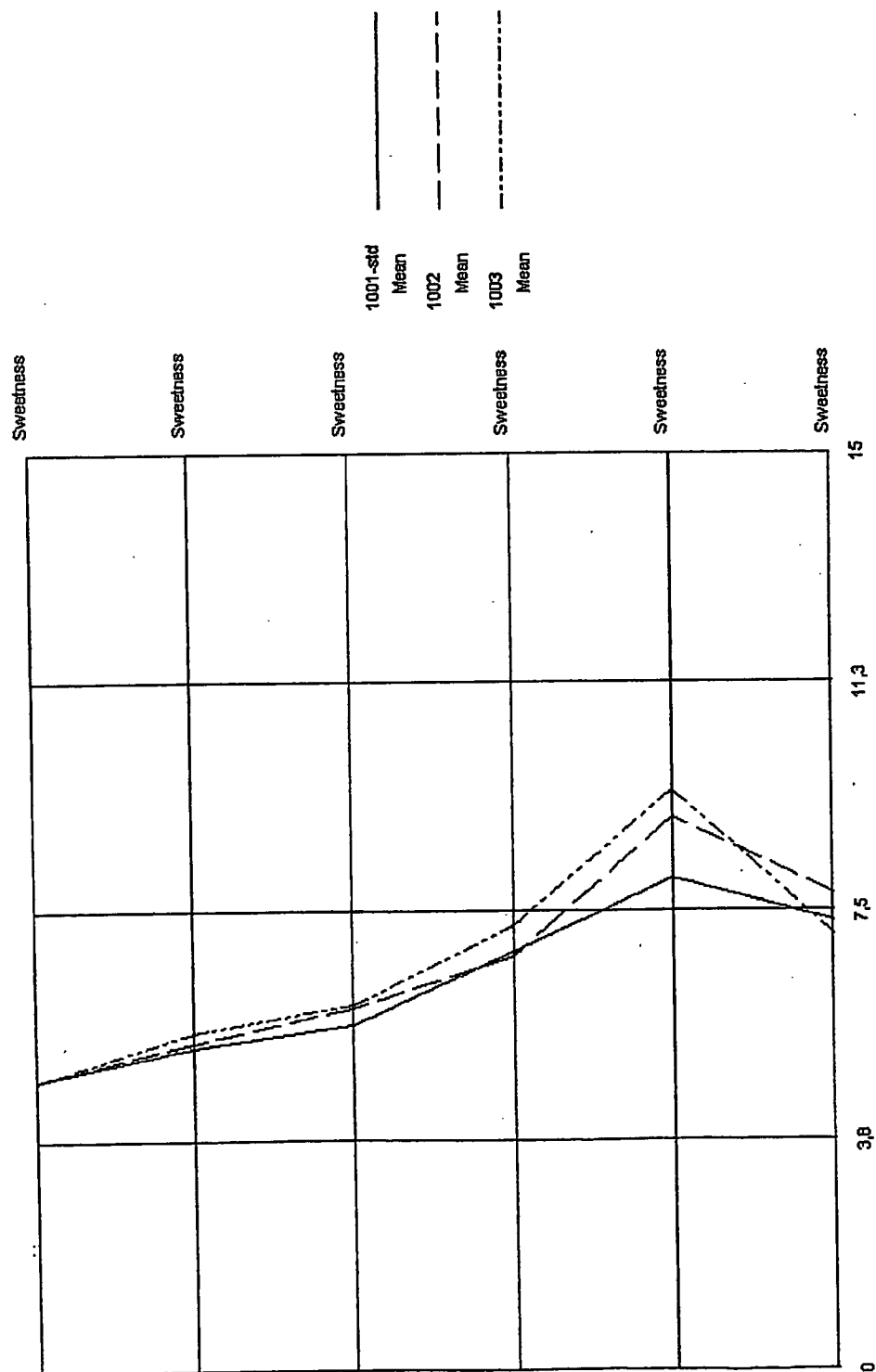


Fig. 13

14 / 16

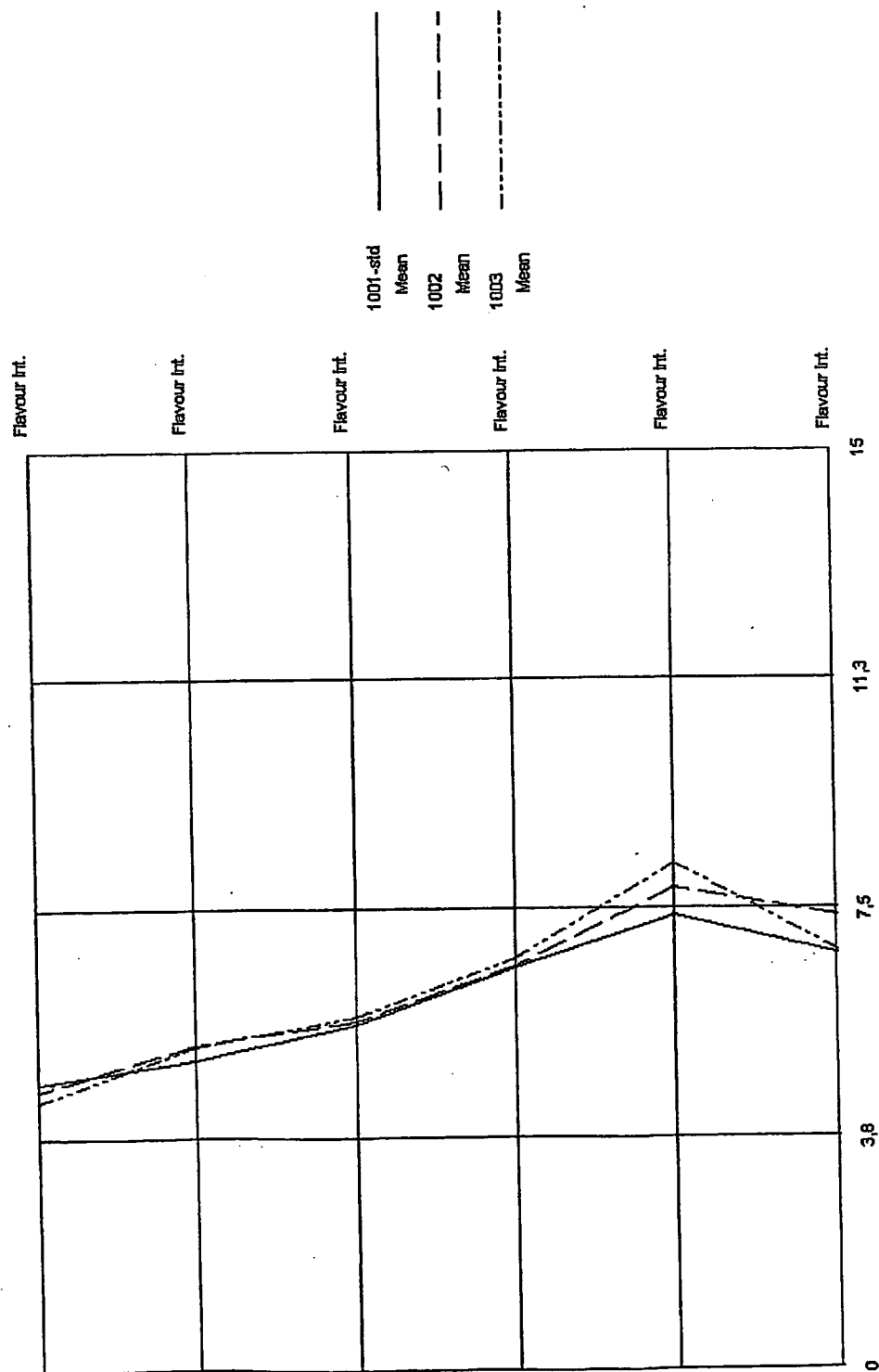


Fig. 14

15 / 16

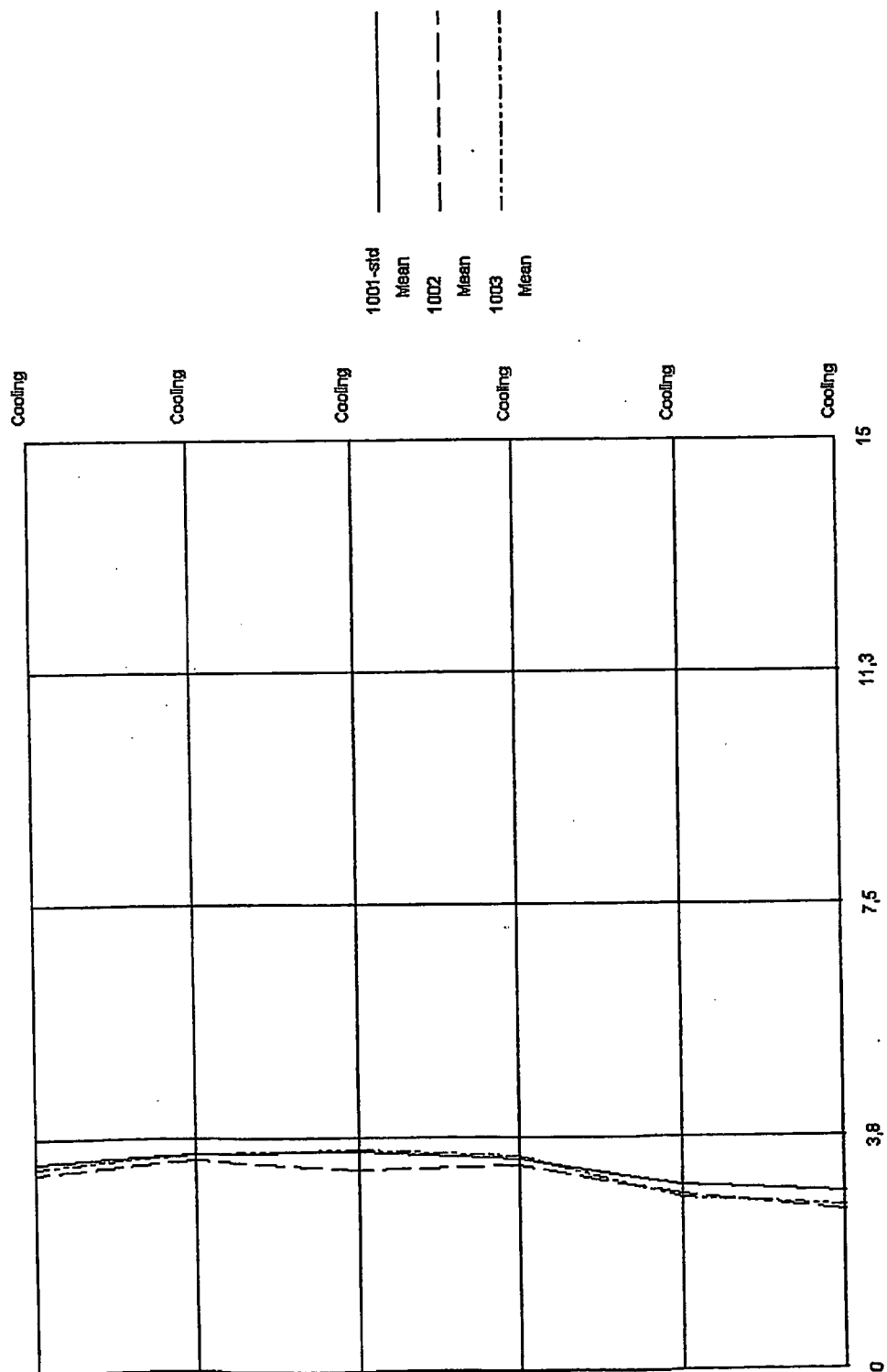


Fig. 15

16 / 16

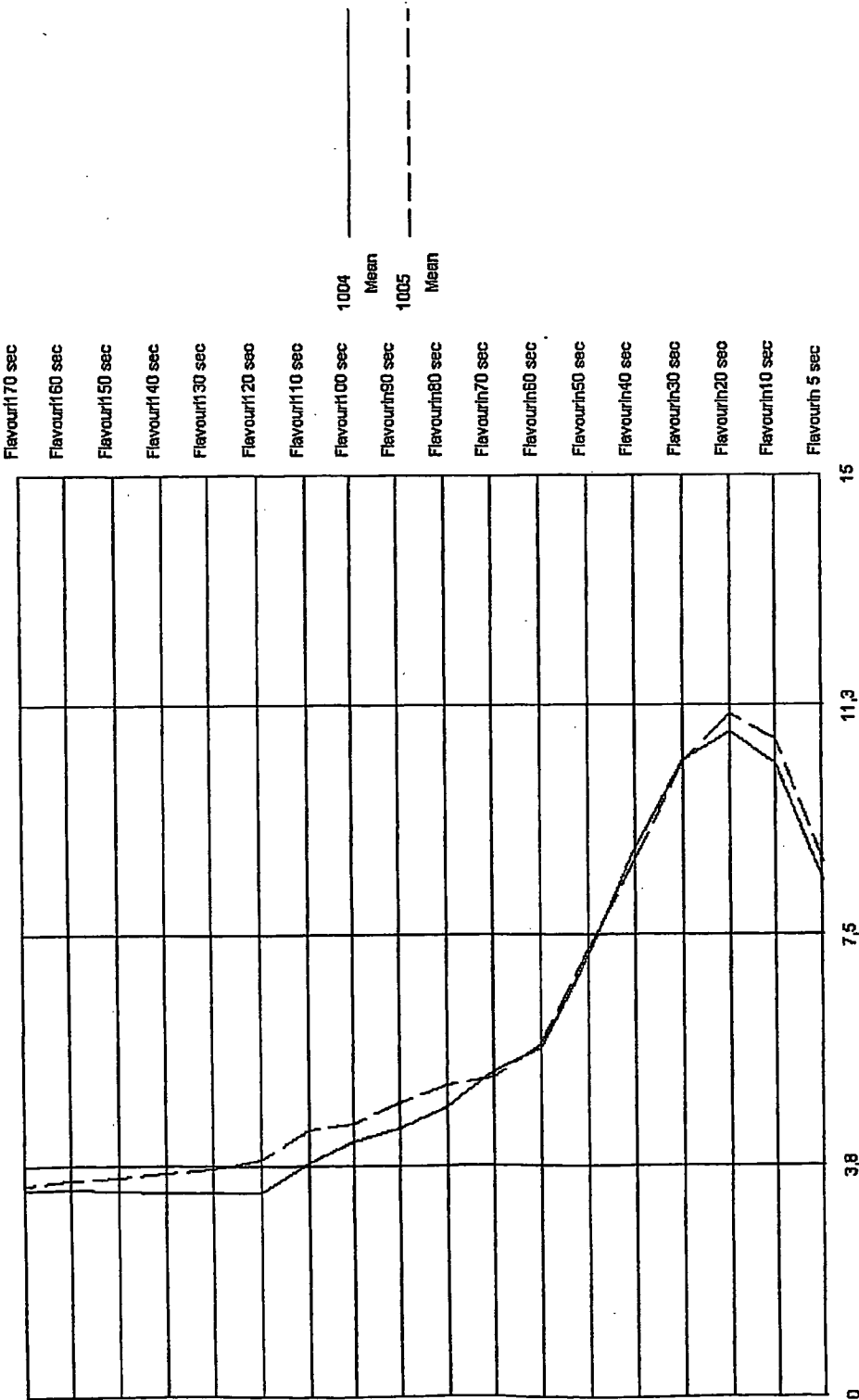


Fig. 16

INTERNATIONAL SEARCH REPORT

International Application No
PCT/DK 02/00628

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 A23G3/30 //C08G63/08,C08G63/64,C08L67/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 A23G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 01 47368 A (PATEL BHARAT KANAIYALAL ;GOLDBERG DANIEL (US); EATON ROBERT FRANCI) 5 July 2001 (2001-07-05) page 17, paragraph 4; claims abstract	1-64
A	US 6 441 126 B1 (TOMLINSON CHARLES R ET AL) 27 August 2002 (2002-08-27) abstract; claims	1-64
A	WO 94 11441 A (UNIV GRONINGEN ;GRIJPM A DIRK WYBE (NL); JOZIASSE CONRNE LIS AARNOUD) 26 May 1994 (1994-05-26) page 8, line 29 - line 33; claims abstract	1-64
	--- -/-	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents:

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- "&" document member of the same patent family

Date of the actual completion of the international search

22 May 2003

Date of mailing of the international search report

12.06.2003

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 851 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

MONIKA BOHLIN / ELY

INTERNATIONAL SEARCH REPORT

International Application No

PCT/DK 02/00628

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 672 367 A (GRIJPMAN DIRK WYBE ET AL) 30 September 1997 (1997-09-30) abstract; claims ---	1-64
E	WO 02 076230 A (ANDERSEN LONE ;DANDY AS (DK); ISAKSEN ANETTE (DK); WITTORFF HELLE) 3 October 2002 (2002-10-03) abstract; claims ---	1-64
E	WO 02 076228 A (ANDERSEN LONE ;DANDY AS (DK); ISAKSEN ANETTE (DK); WITTORFF HELLE) 3 October 2002 (2002-10-03) abstract; claims ---	1-64
E	EP 1 306 013 A (HYCAIL B V) 2 May 2003 (2003-05-02) the whole document -----	1-64

INTERNATIONAL SEARCH REPORT

information on patent family members

International Application No

PCT/DK 02/00628

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
WO 0147368	A	05-07-2001	AU 2608701 A WO 0147368 A1	09-07-2001 05-07-2001
US 6441126	B1	27-08-2002	US 2003032764 A1 US 6469129 B1 US 6444782 B1	13-02-2003 22-10-2002 03-09-2002
WO 9411441	A	26-05-1994	NL 9201949 A AU 5577894 A DE 69312312 D1 DE 69312312 T2 EP 0667885 A1 JP 3328285 B2 JP 8504850 T WO 9411441 A1	01-06-1994 08-06-1994 21-08-1997 08-01-1998 23-08-1995 24-09-2002 28-05-1996 26-05-1994
US 5672367	A	30-09-1997	NL 9401703 A EP 0711506 A2 FI 954867 A JP 8196214 A	01-05-1996 15-05-1996 15-04-1996 06-08-1996
WO 02076230	A	03-10-2002	WO 02076230 A1 WO 02076231 A1	03-10-2002 03-10-2002
WO 02076228	A	03-10-2002	WO 02076228 A1 WO 02076231 A1	03-10-2002 03-10-2002
EP 1306013	A	02-05-2003	EP 1306013 A1 WO 02076232 A1	02-05-2003 03-10-2002

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☒ **BLACK BORDERS**
- ☐ **IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- ☒ **FADED TEXT OR DRAWING**
- ☐ **BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- ☐ **SKEWED/SLANTED IMAGES**
- ☐ **COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- ☐ **GRAY SCALE DOCUMENTS**
- ☐ **LINES OR MARKS ON ORIGINAL DOCUMENT**
- ☐ **REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- ☐ **OTHER:** _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.